X-Ray Laue Diffraction Microscopy

Nano- and microscale X-ray characterisation of functional materials and microstructure

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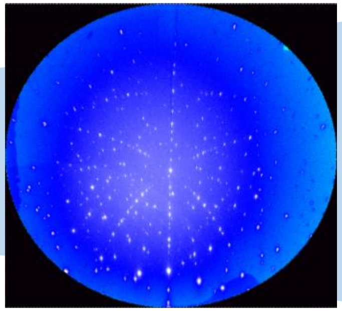
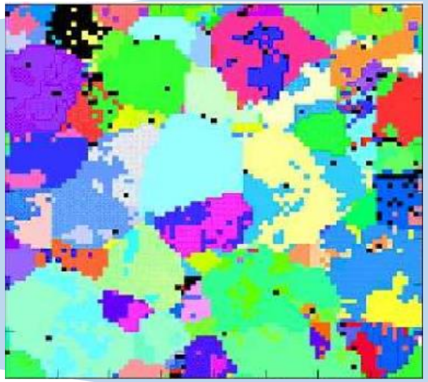
Univ. Grenoble Alpes, CEA/IRIG/MEM & SyMMES, CNRS Institut Néel, CNRS SERAS

CRG-IF BM32 beamline @ ESRF



Single crystal Laue Pattern



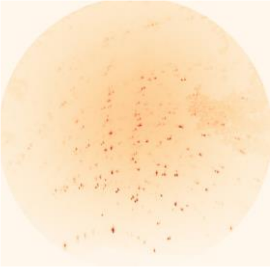
2D map:
Orientation
dev. strain map

Complex materials
Polycrystals, defects

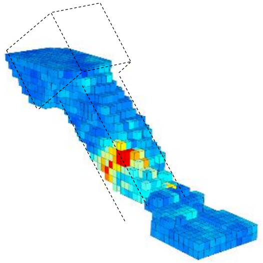
3D map

Full tensor stress/strain

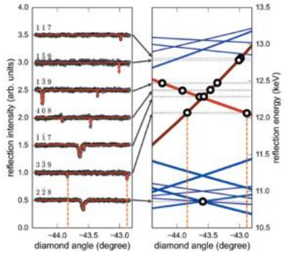
AI-based Indexing



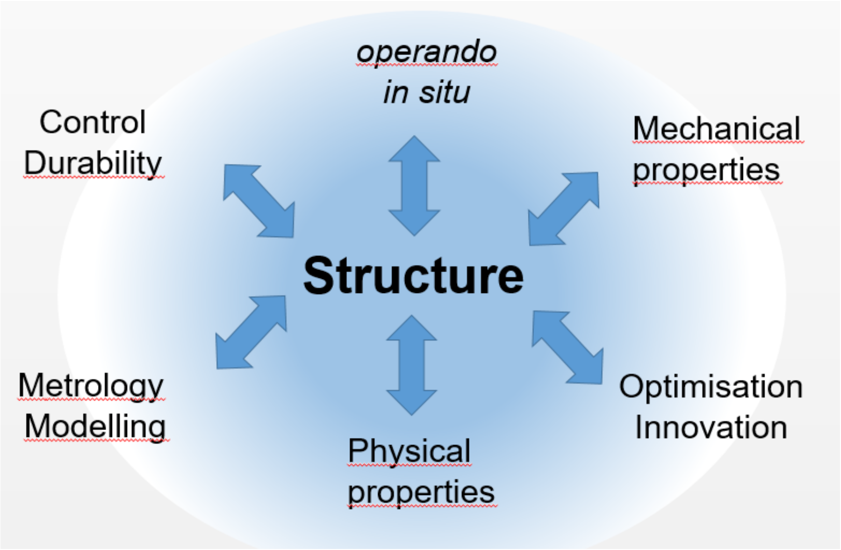
Depth resolution



Energy resolution



Objectives & scopes

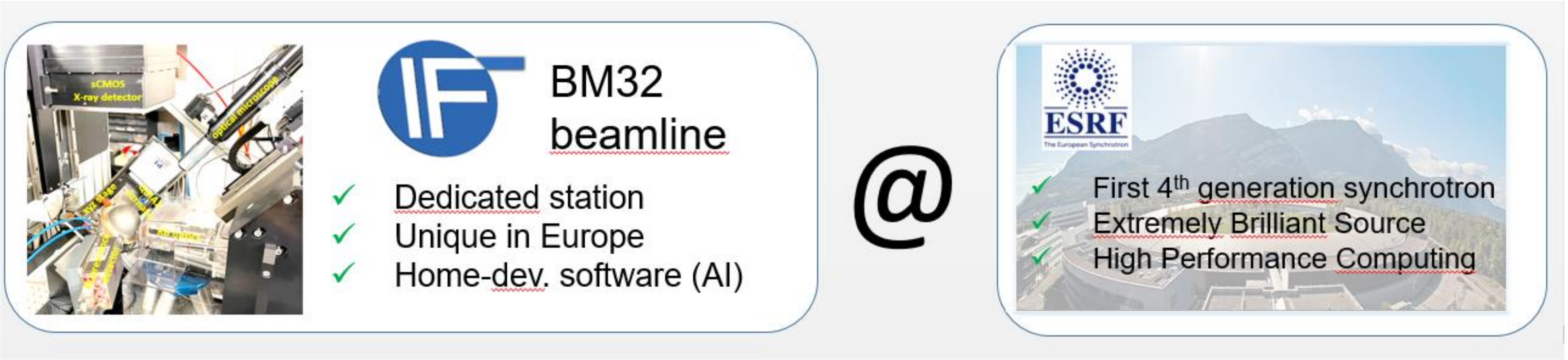


For

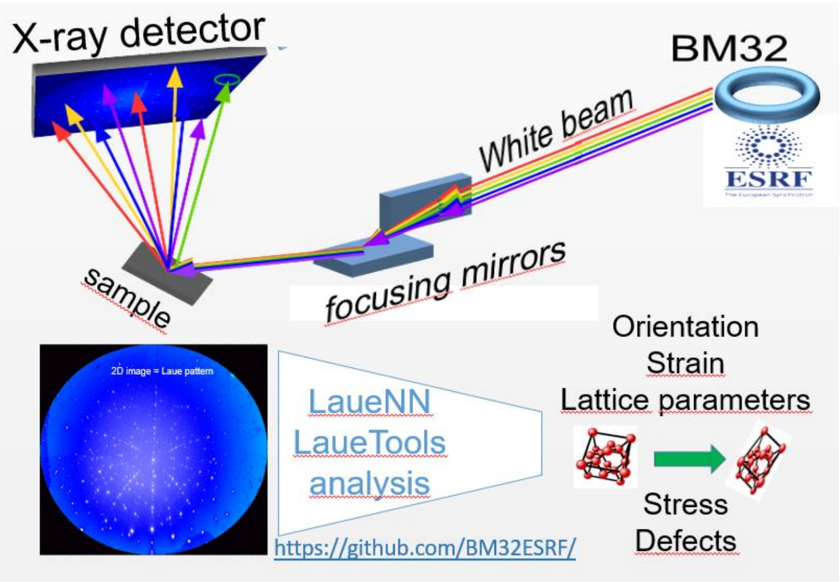
Materials science, Nanoscience
Real and model systems
Microstructure, nano- and microobjects, microdevices
Fundamental science & technology-related materials

Needs

Accurate, quantitative experimental data
Accessible measurements
Transversal technique (metals, oxide, semiconductors,…)
Complementary with SEM, TEM



Laue Microscopy

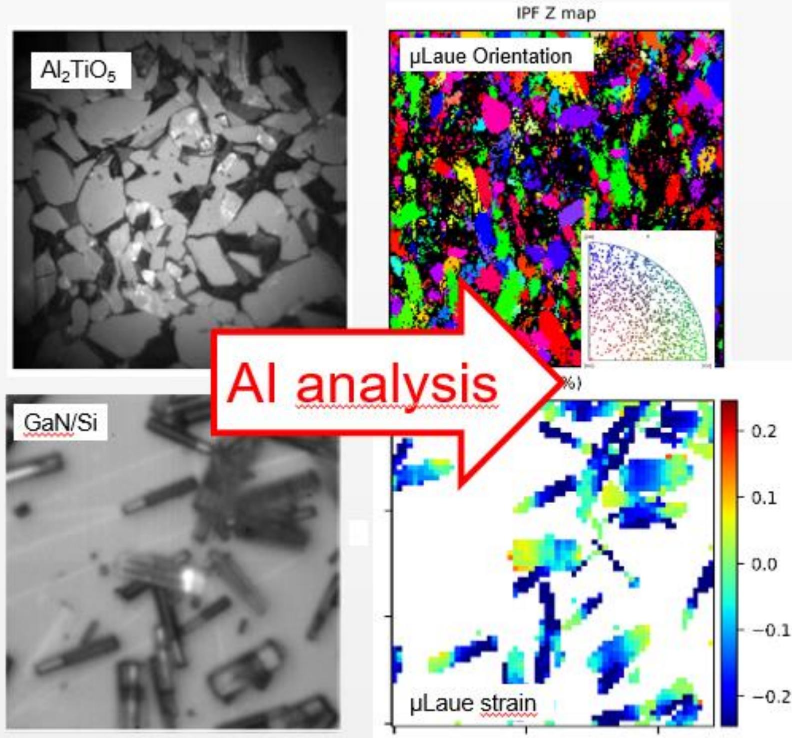


Local lattice parameters, strain, orientation, microstructure

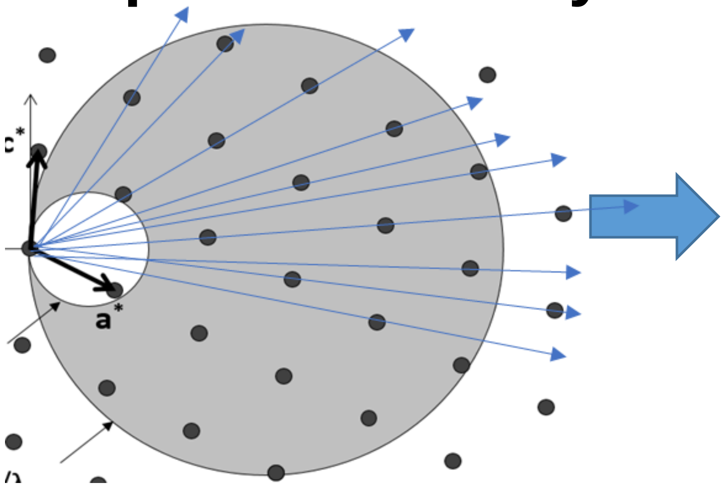
HR spatial: 300 nm x 300 nm
HR angular: 0,006° ⇔ 0,01%

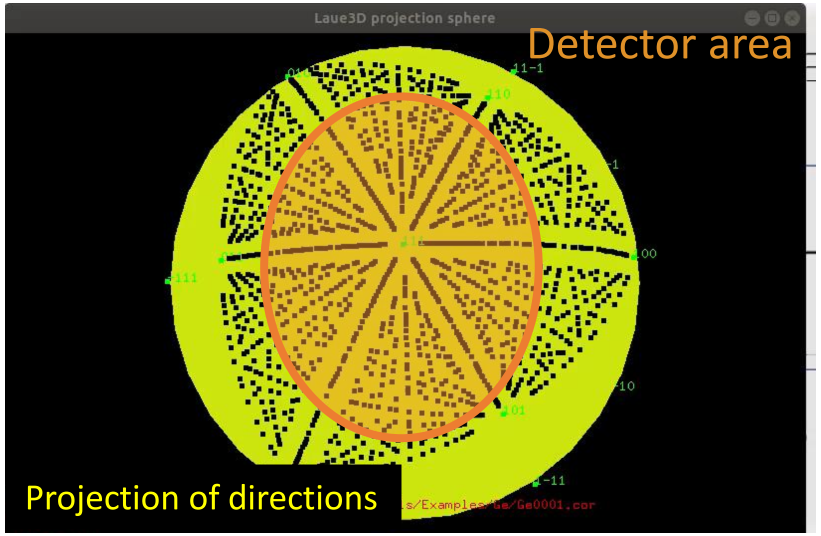
Capabilities
No sample preparation
In situ & operando (T, force, I, V, XEOL,…)
Polycrystalline & single crystal
2D mapping (9000 pts/h) Flexible FOV
Damaged material

Augmented µLaue
Depth resolution (500nm)
Stress assessment
Extended defects (plasticity)

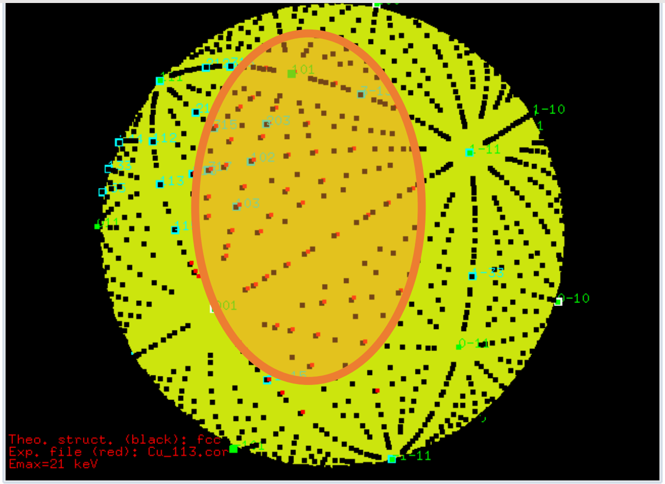


Principles: sensitivity & resolution

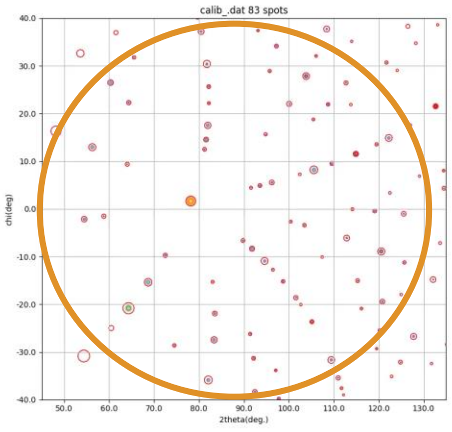




Absolute positions => orientation



Relative positions / reference => strain



Effect of strain or lattice parameters change

Standard Laue diffraction

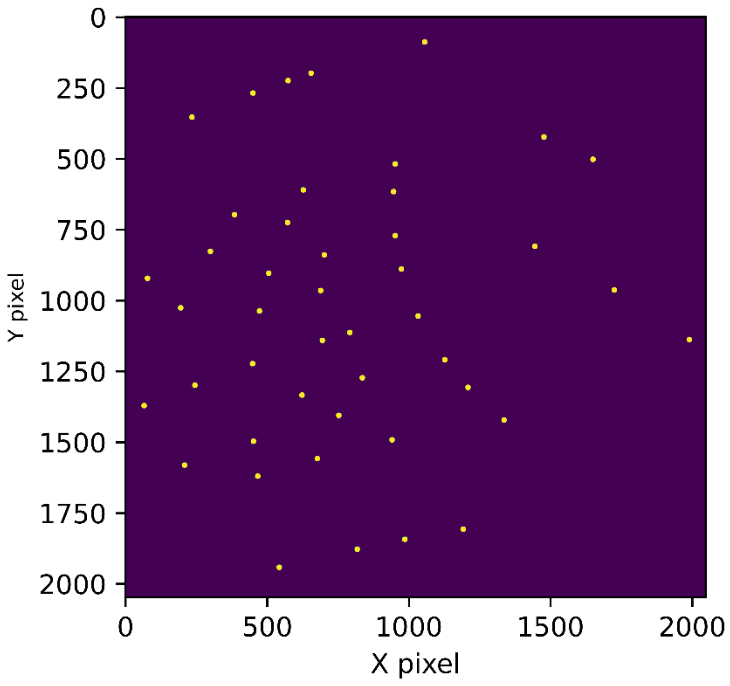
Experimental & quantitative determination of unit cell :

- Orientation
- (angular) Shape
  (lattice parameters, strain)

- **Absolute** values
- **10^{-4} resolution** for
  - high absorbing material
  - low defects density
- **Better resolution on gradients**

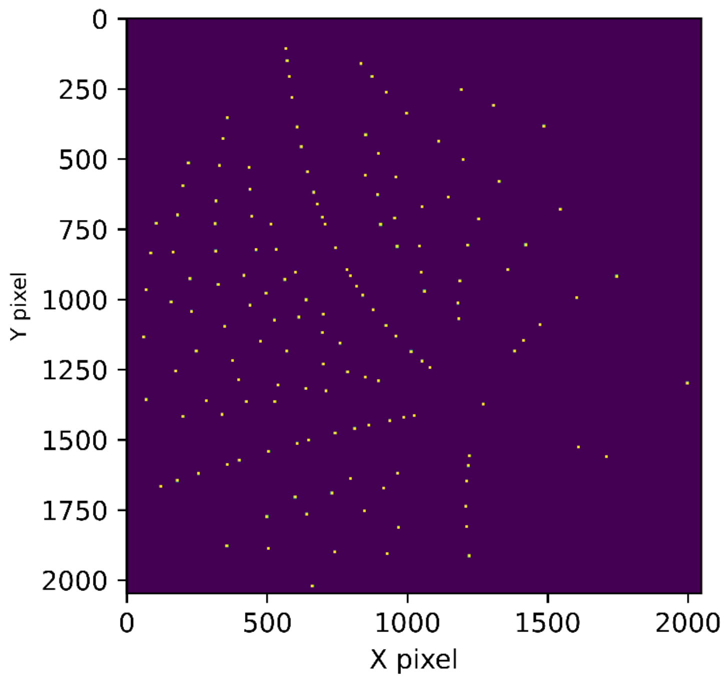
AI-based Laue pattern indexing: complexity

Simulated “complete” Laue pattern
Single crystal Cu
(cubic)



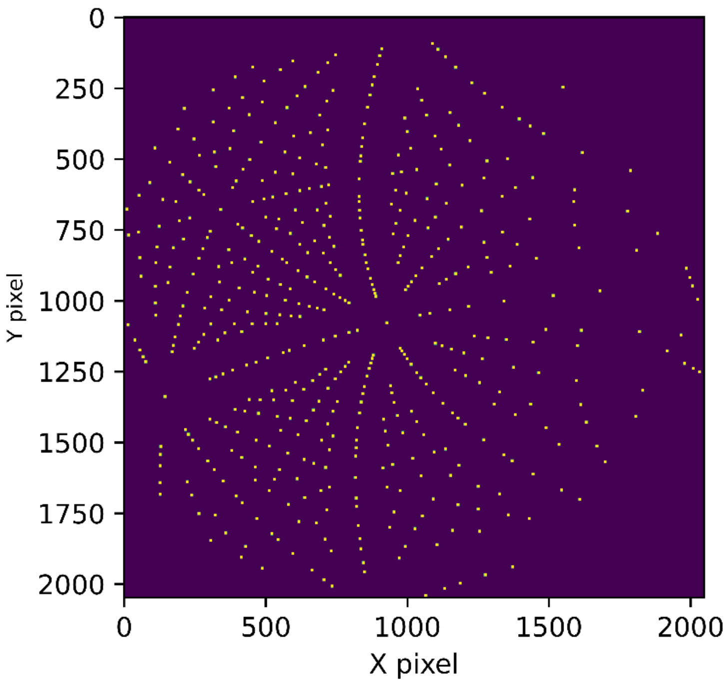
Nb. of spots in detector: ~40

Simulated “complete” Laue pattern
Single crystal UO2
(Large cubic unit cell)



Nb. of spots in detector: ~160

Simulated “complete” Laue pattern
Single crystal ZrO2
(monoclinic)

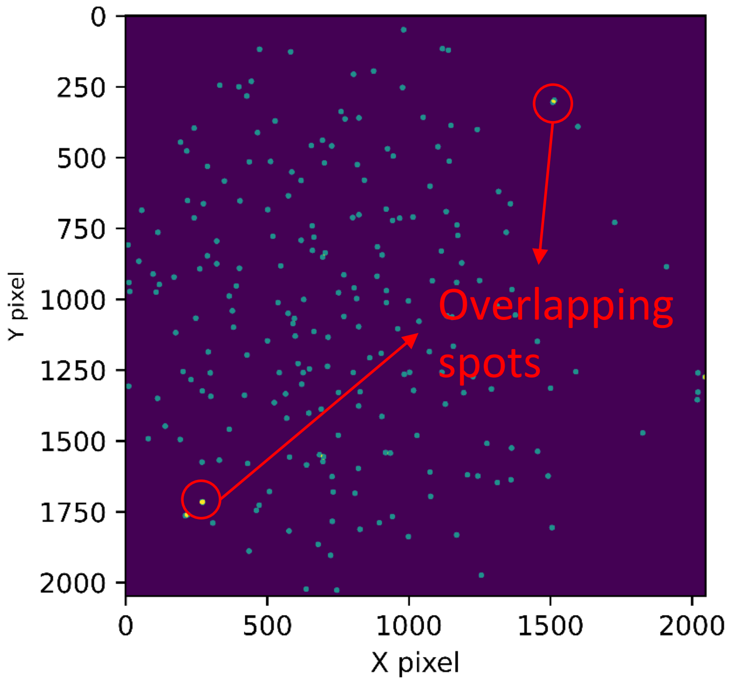


Nb. of spots in detector: ~500

*Detector at 70mm from the sample surface

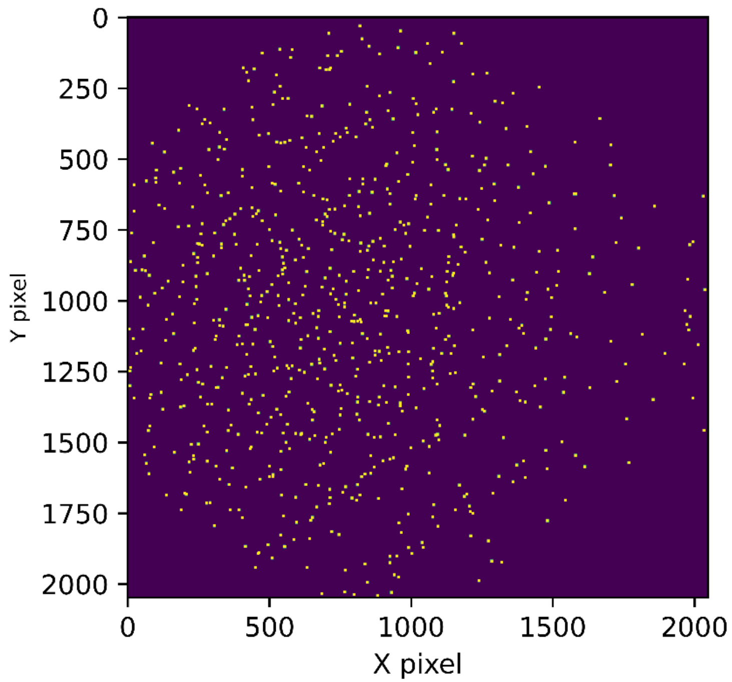
AI-based Laue pattern indexing: complexity

Simulated “complete” Laue pattern
Polycrystalline Cu (5 grains)
(cubic)



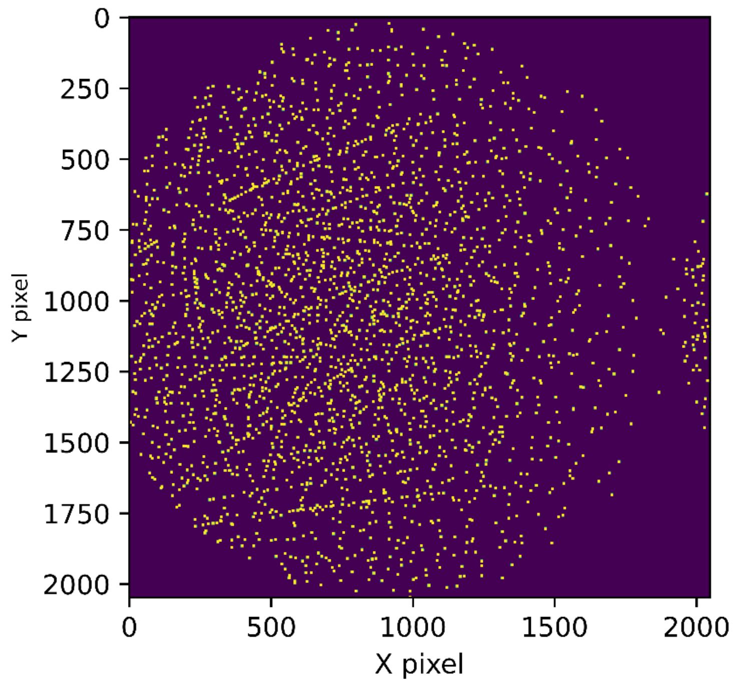
Nb. of spots in detector: ~230

Simulated “complete” Laue pattern
Polycrystalline UO2 (5 grains)
(Large cubic unit cell)



Nb. of spots in detector: ~800

Simulated “complete” Laue pattern
Polycrystalline ZrO2 (5 grains)
(monoclinic)



Nb. of spots in detector: ~2600

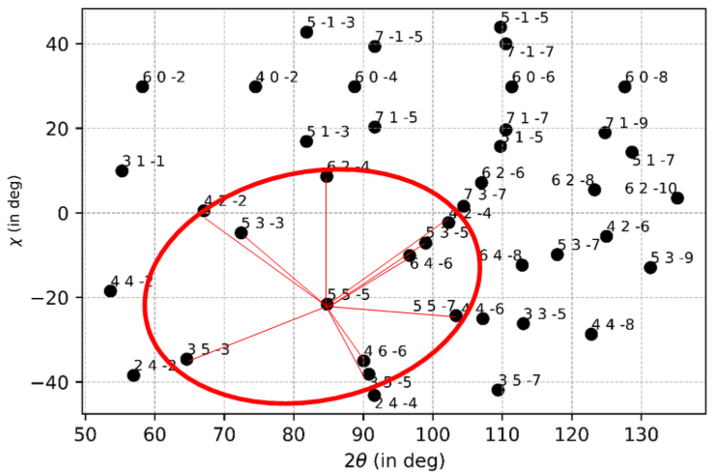
- In reality, we also have photon noise/ dead pixels/ saturated peaks on the detector (also detector efficiency)
- Also the intensity of Laue spots for grains diffracting underneath the surface grains will not be the same

AI-based Laue pattern indexing: extracting Laue features for training

Ability of the neural network learning depends strongly on the applicability of the feature it is dealt with.

Angular space is more meaningful in Laue

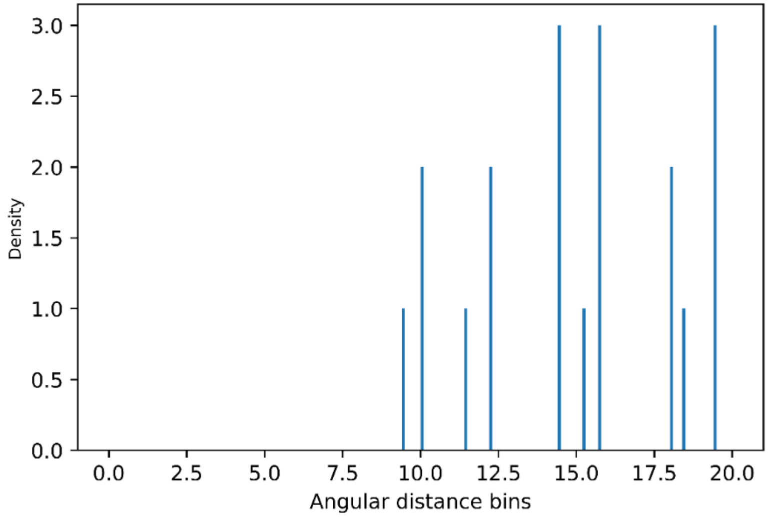
Simulated Laue Pattern for single crystal Cu



Binning of angular distance between neighbor spots

Neighbors defined by limit search angle (here 20°)

Angular distribution for (55-5)

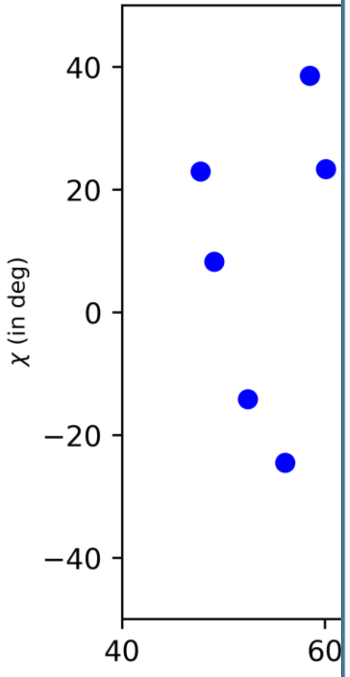


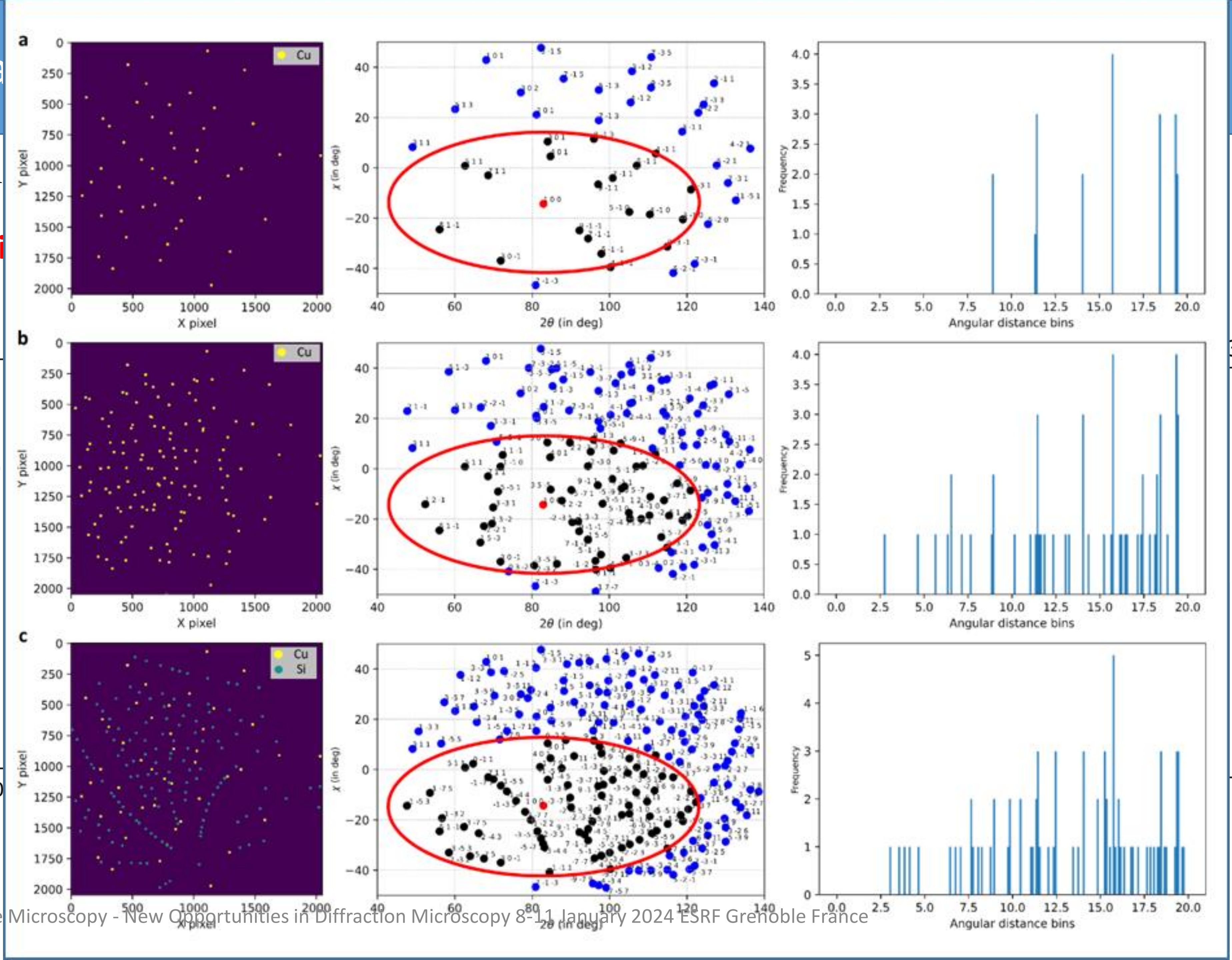
→ **Angular distance distribution is used as input for the Neural network.**

AI-based La

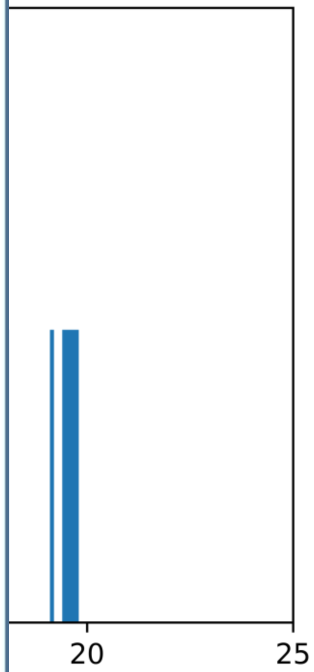
Ability of the neu

Angular space i



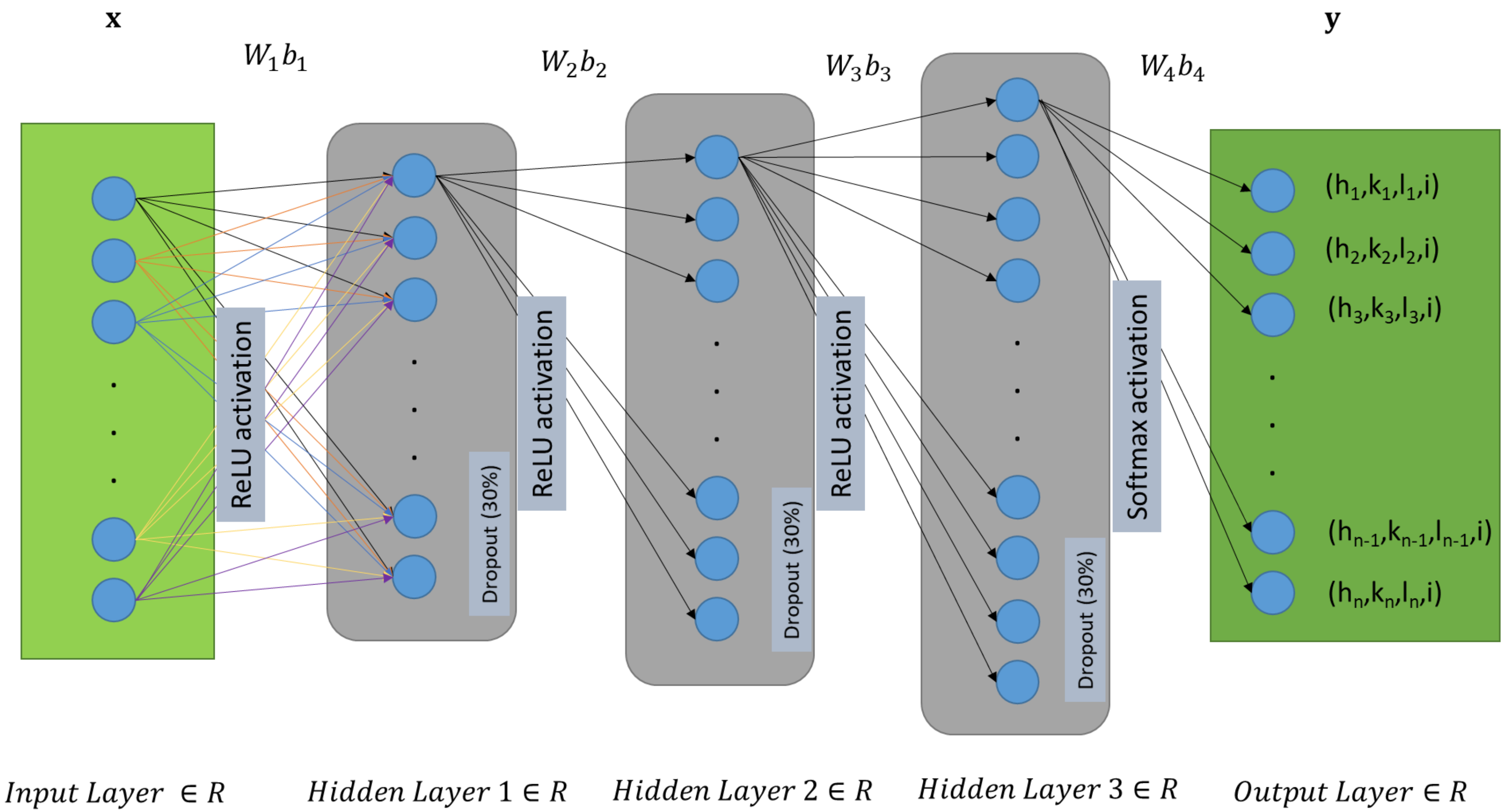


3)



AI-based Laue pattern indexing : an optimized Deep Feed Forward model

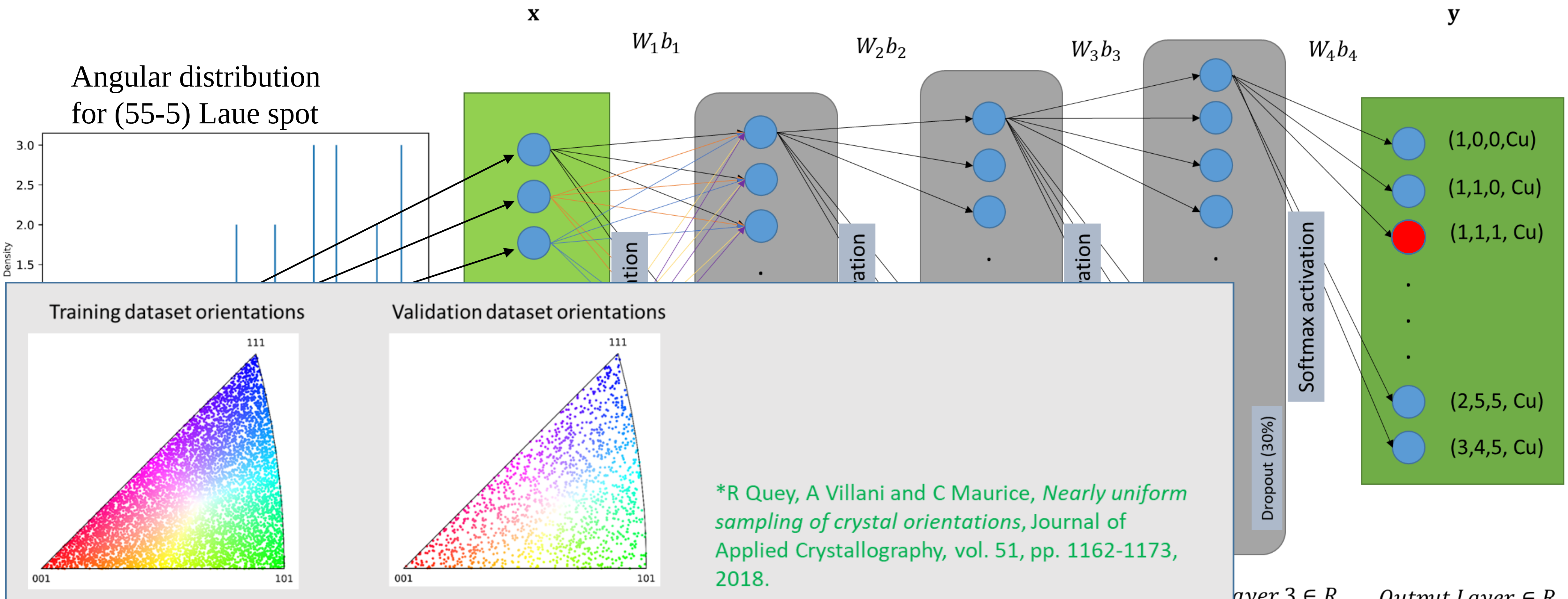
A simple NN architecture → Faster prediction



*Purushottam raj purohit R.R.P, et al. "LaueNN: Neural network based *hkl* recognition of Laue spots and its application to polycrystalline materials". ." Journal of Applied Crystallography 55, no. 4 (2022).

AI-based Laue pattern indexing : an optimized Deep Feed Forward model

A simple NN architecture → Faster prediction



Uniform orientation distribution generated with Neper*

*R Quey, A Villani and C Maurice, *Nearly uniform sampling of crystal orientations*, Journal of Applied Crystallography, vol. 51, pp. 1162-1173, 2018.

- Data augmentation: Gaussian noise and disappearance of spots (or **partial Laue patterns**) based on their energies

*Purushottam raj purohit R.R.P, et al. "LaueNN: Neural network based *hkl* recognition of Laue spots and its application to polycrystalline materials". ." Journal of Applied Crystallography 55, no. 4 (2022).

AI-based Laue pattern indexing : an optimized Deep Feed Forward model

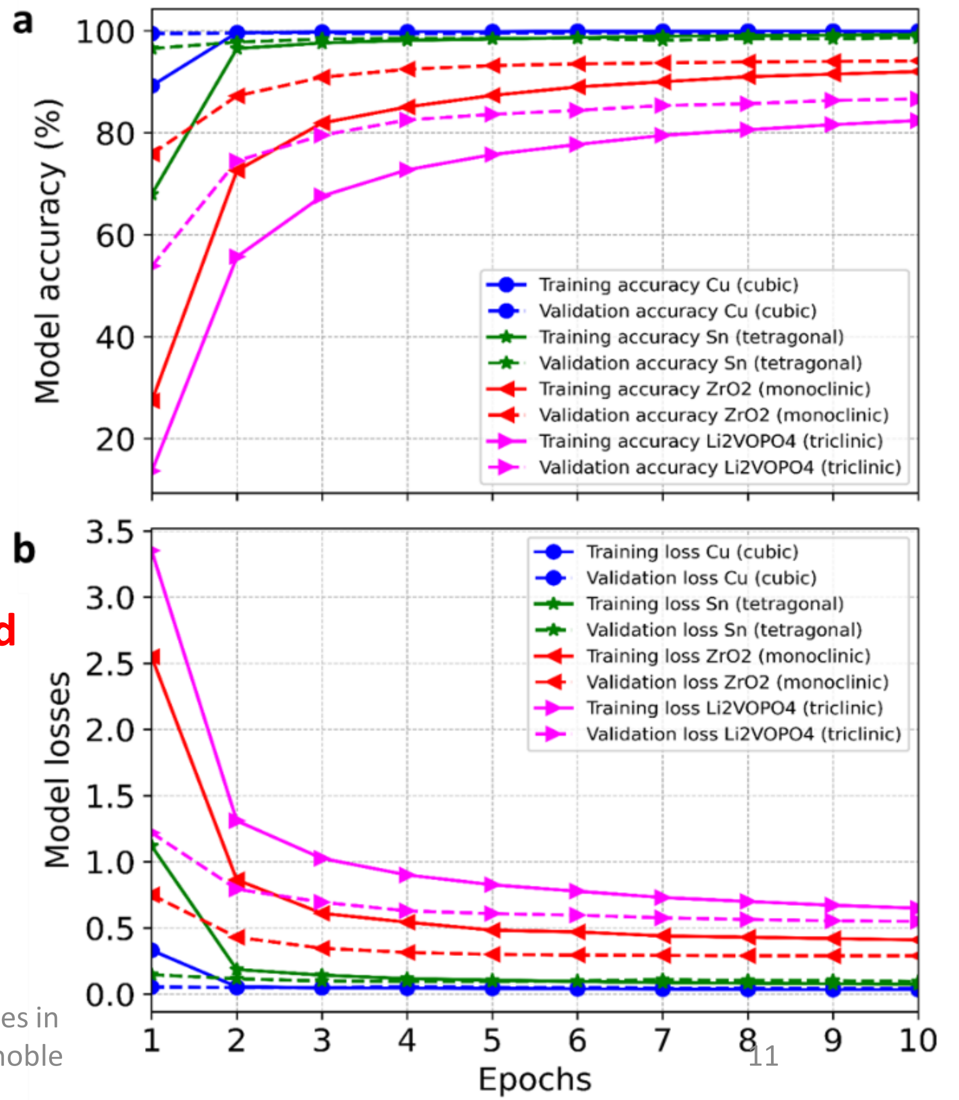
Compound	Crystal system	Model training time (s)	Validation accuracy of the model	Time to predict hkls in one Laue image (s)
Cu	Cubic	25	99.7%	0.25
Ti	Hexagonal	180	98.2%	0.33
Sn	Tetragonal	180	98.6%	0.40
ZrO_2	Monoclinic	750	94.4%	0.65
Li_2VOPO_4	Triclinic	1200	88.2%	0.86

<1 second

Training: Standard laptop with 8cores
Prediction: Single CPU mode

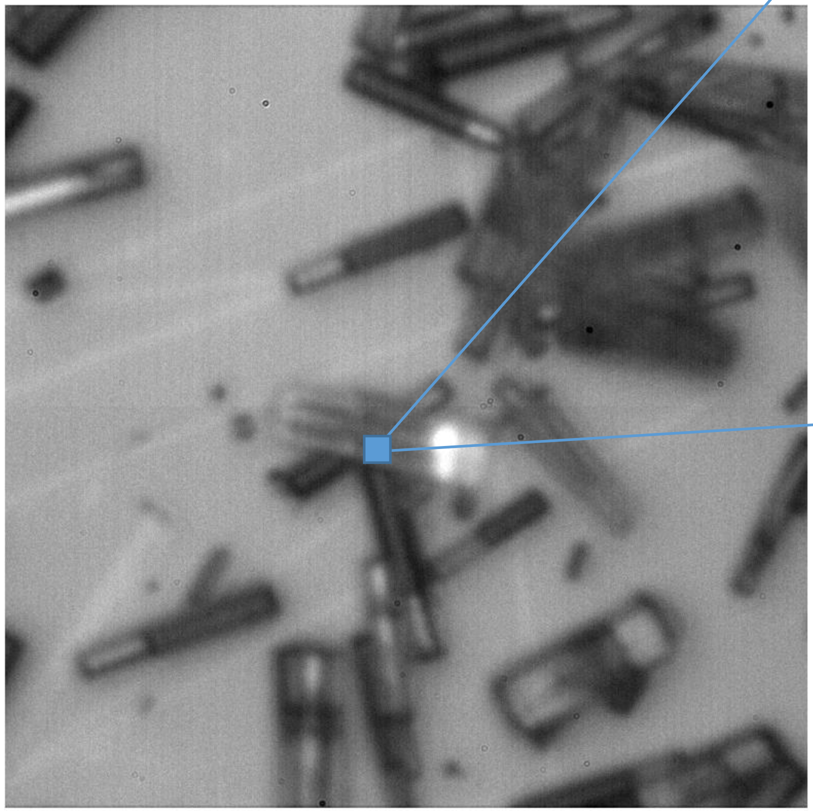
Available Lauetools, LaueNN on github

- Single neural network architecture that works for all crystal symmetries



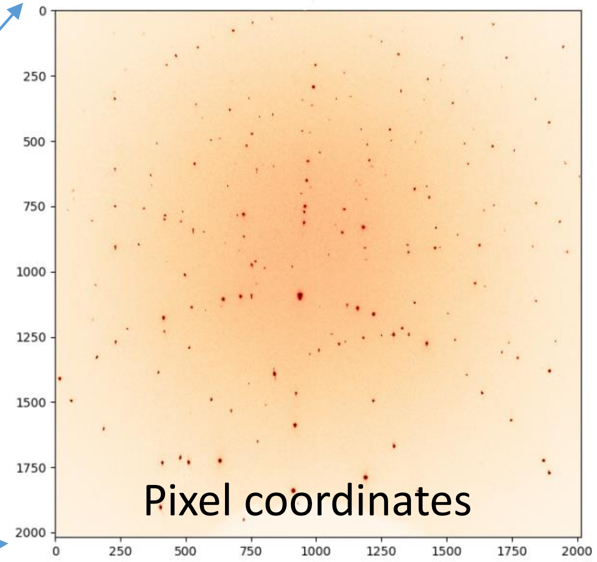
GaN NWs deposited on Si subtrate

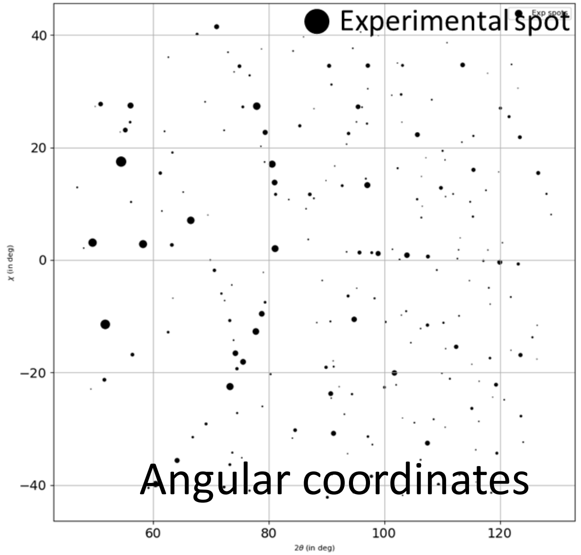
Optical microscopy image



Scan direction

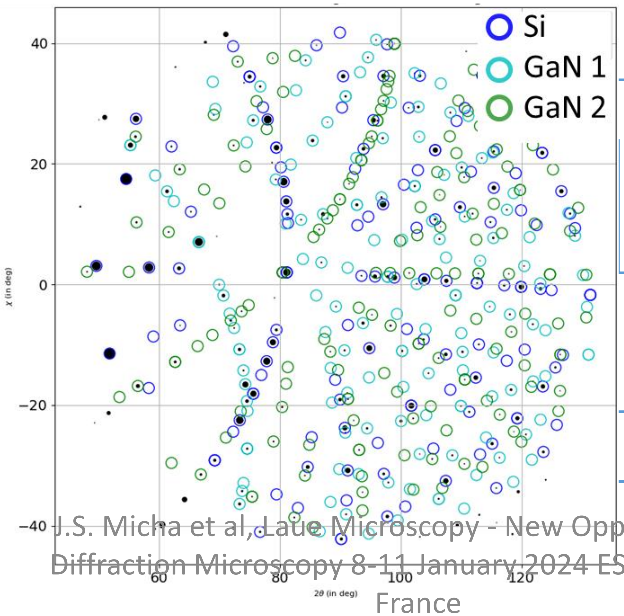
Micro-Laue campaign of Joël Eymery
Univ. Grenoble Alpes, CEA, IRIG-MEM, Nanostru
and Synchrotron Radiation Laboratory.

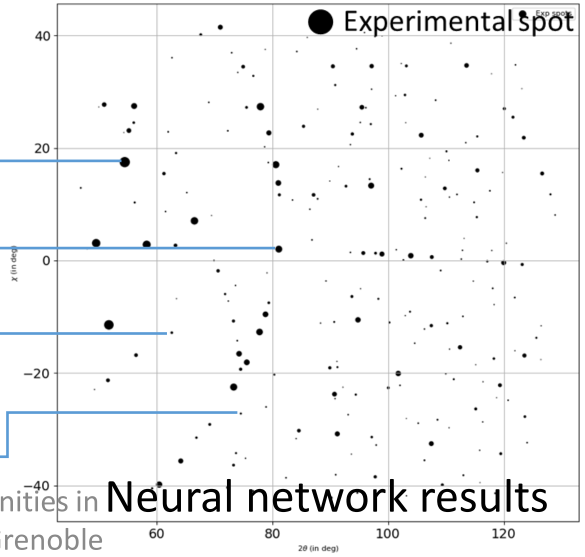




Preprocess
(build histograms)

FFNN

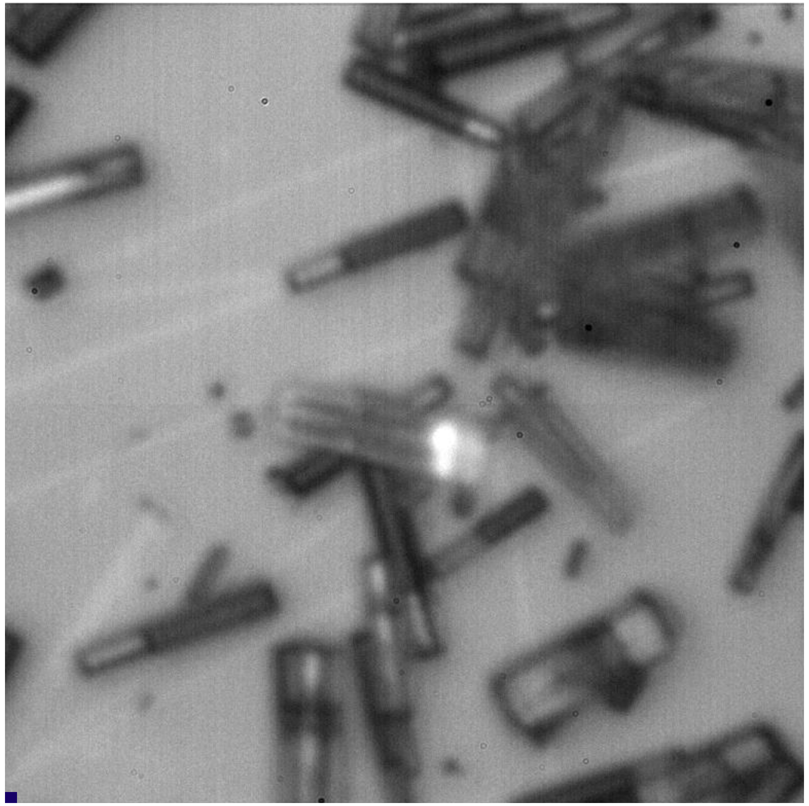




GaN NWs deposited on Si subtrate

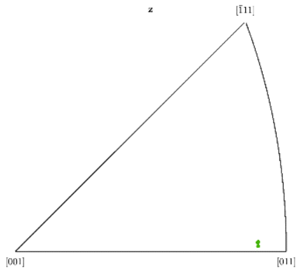
*IPF (Z) plotted with MTEX

Optical microscopy image

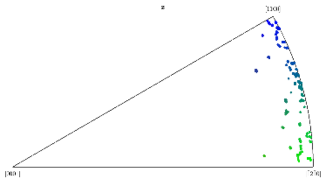


Scan direction

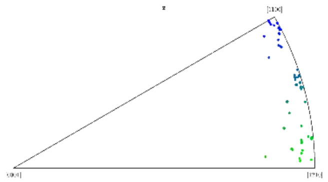
Si- phase



GaN- phase (grain 1)

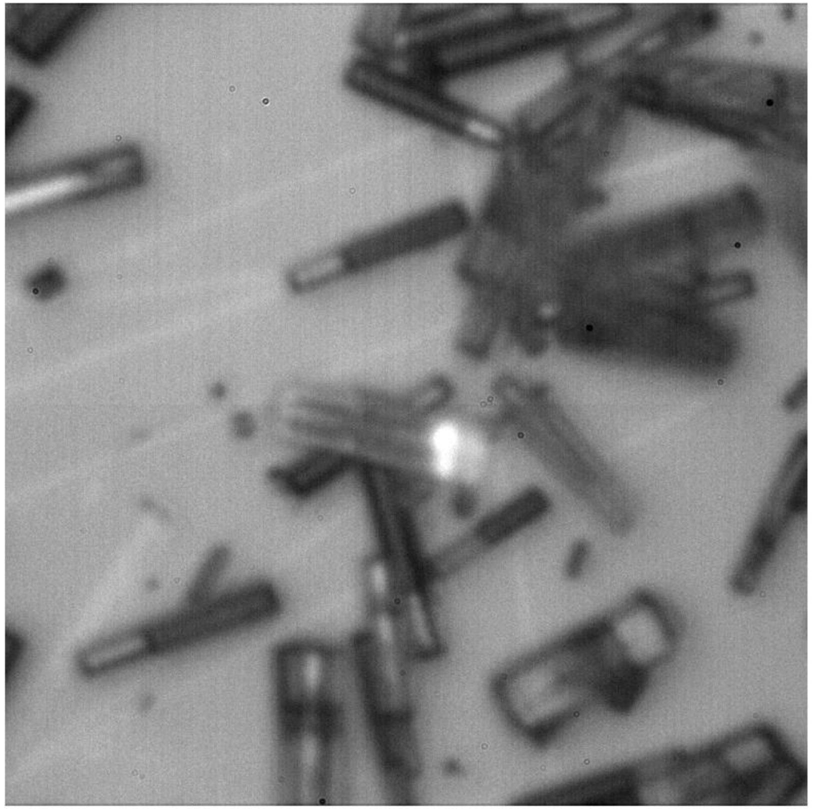


GaN- phase (grain 2)

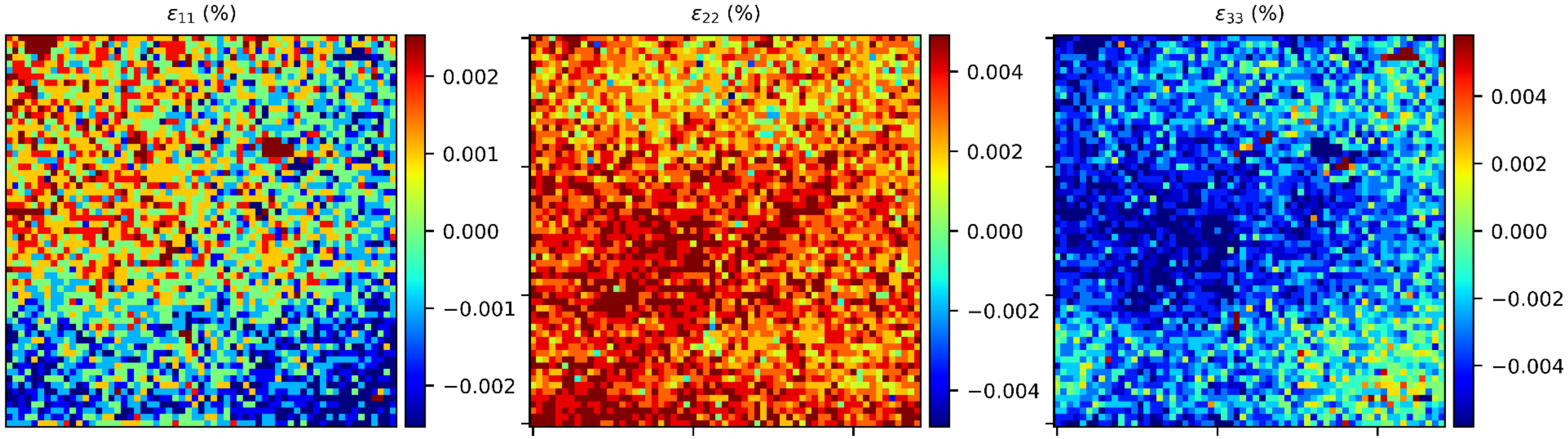


GaN NWs deposited on Si subtrate

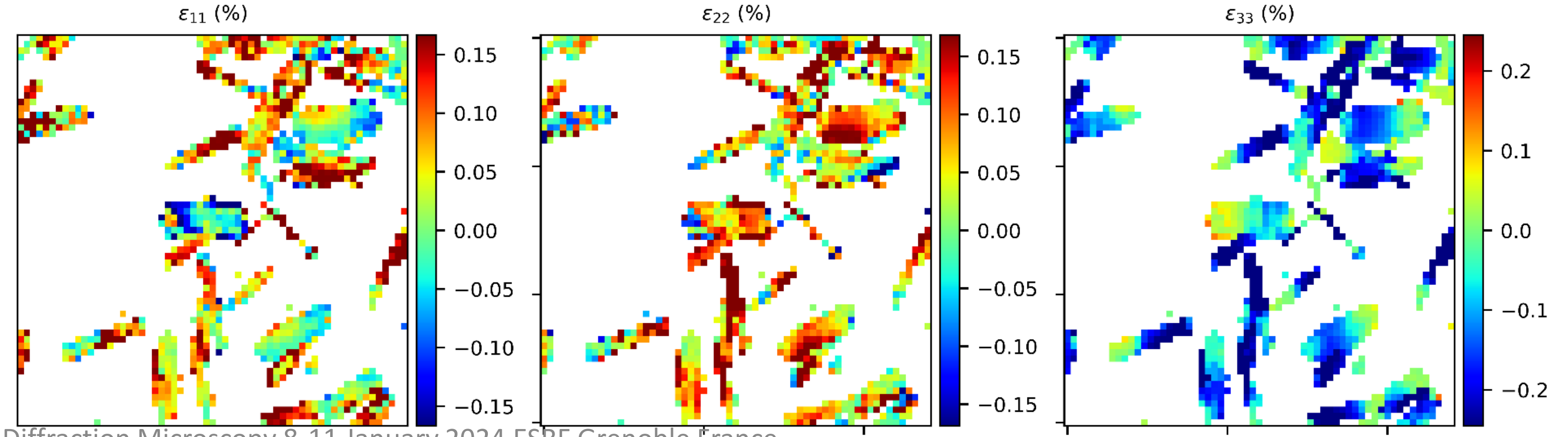
Optical microscopy image



Si- phase

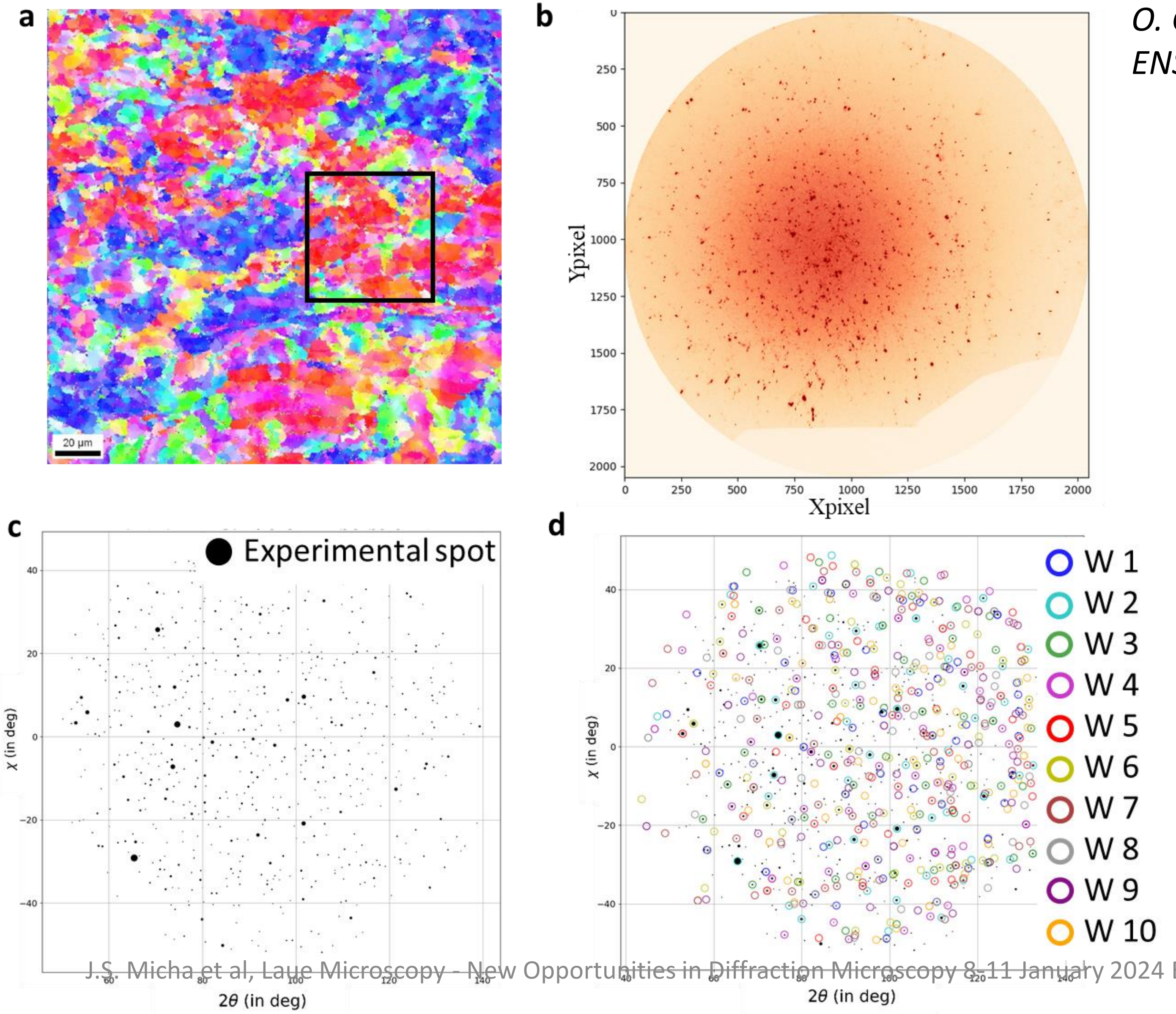


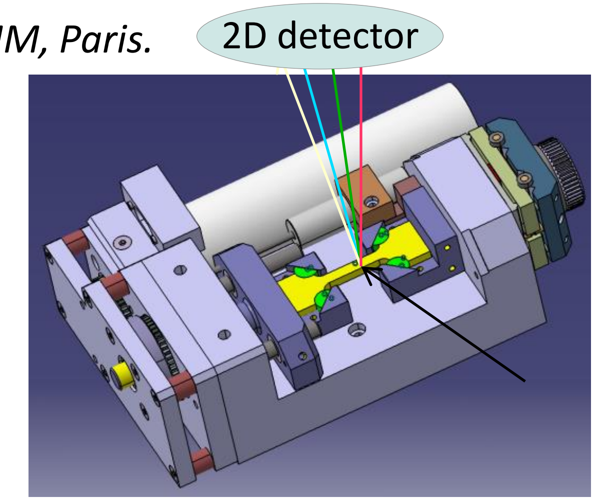
GaN- phase (grain 1)



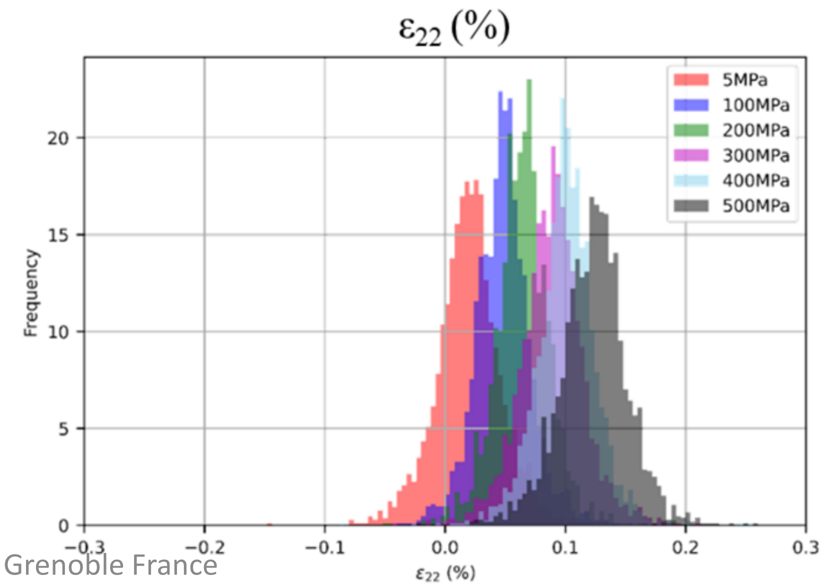
Polycrystalline Tungsten: validation of strain assessment

O. CASTELNAU et al
ENSAM ParisTech/PIMM, Paris.





Tensile test



Key step for high resolution: geometry calibration

High strain resolution obtained by a fine calibration of the geometry
⇔ accurate location of point C wrt 2D detector pixels plane array

Use of optical microscope focusing plane

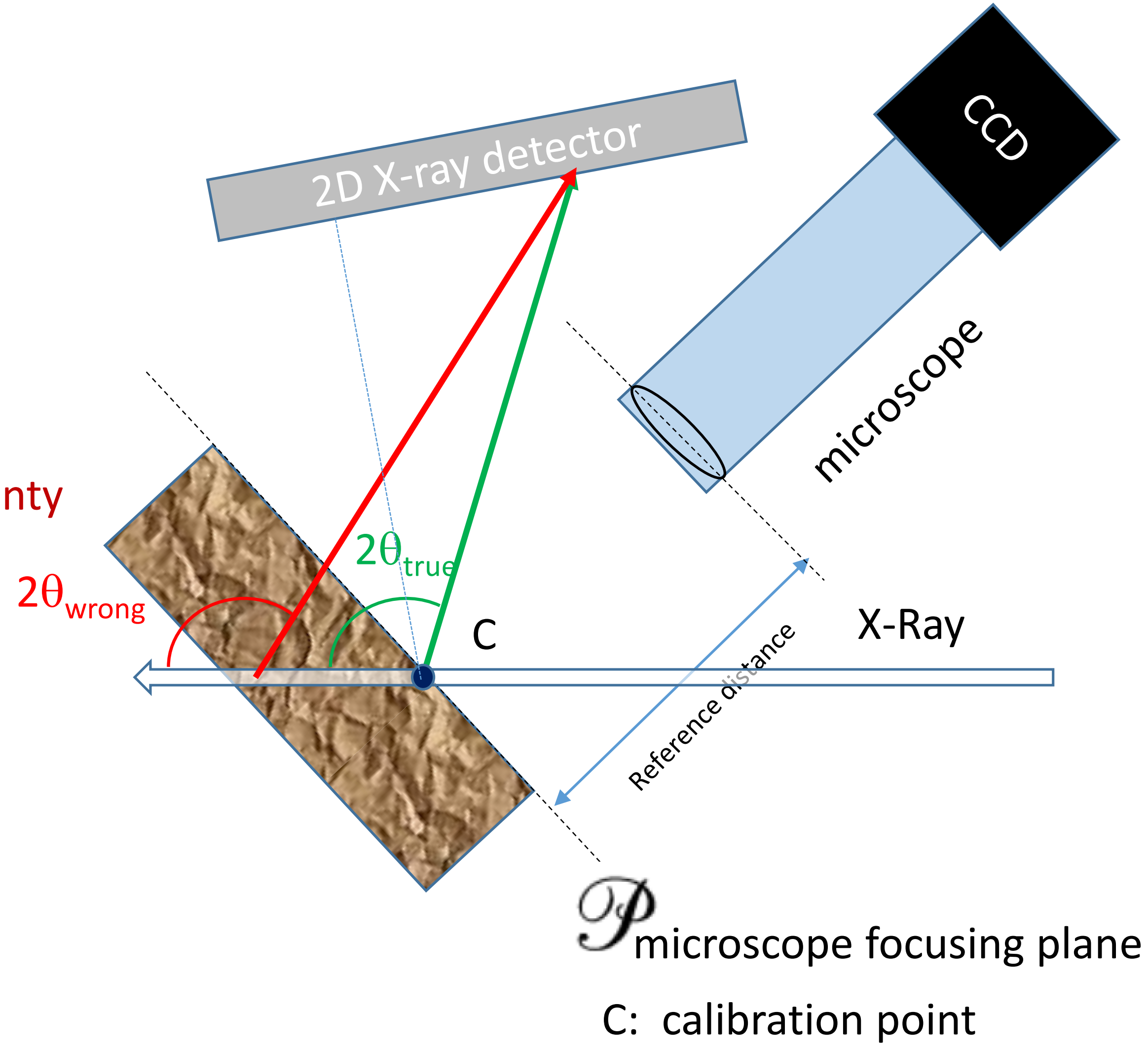
10^{-4} resolution on scattering angles,
absolute strain and absolute lattice parameters
if scattering is close to point C within 10 µm along the beam

Degraded resolution if depth parallax: 10^{-3} for 75 µm depth uncertainty

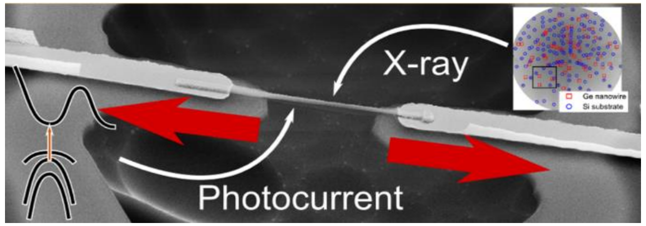
10^{-4} resolution for:

- Objects at surface:
  - High absorbing material
  - Thin films
- Controlled depth of scattering process:
  - Prior knowledge (multilayer, …)
  - Experimentally determined (3D Laue)

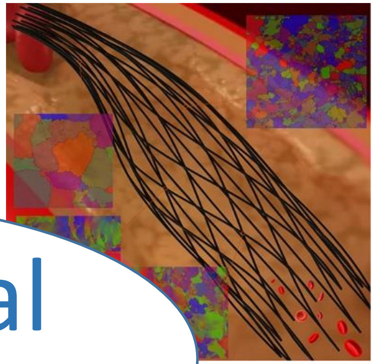
Better resolution on strain gradient than on absolute strain



Applications

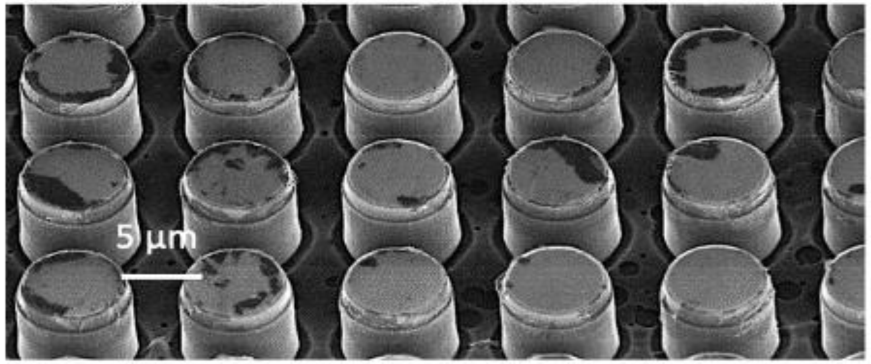


Direct gap strained Ge

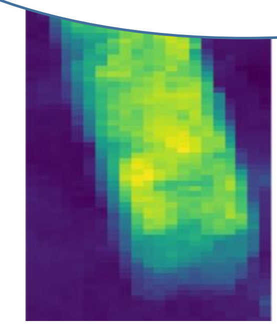


NiTi alloy

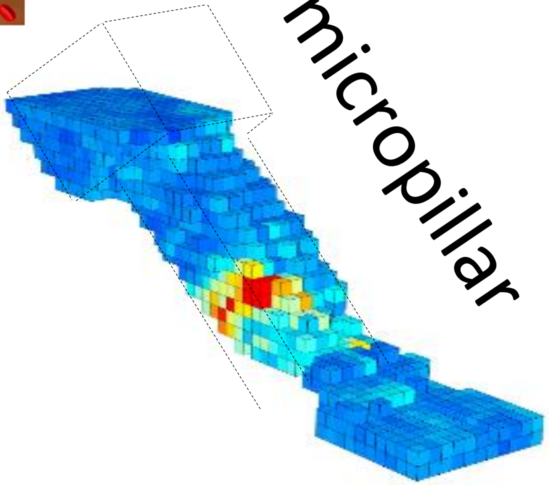
InGaN/GaN µLEDs



Functional materials



ZrO_2
at 1350 °C

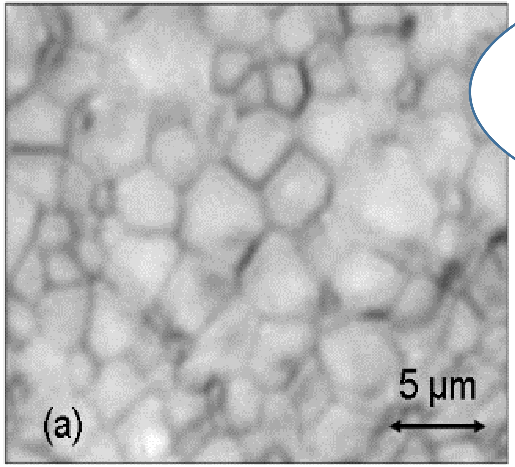


micropillar

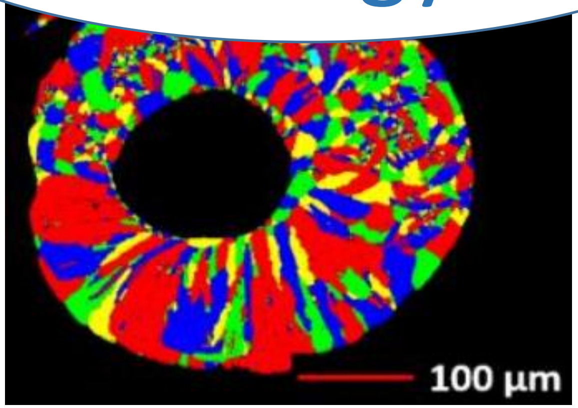
In situ !

Applications

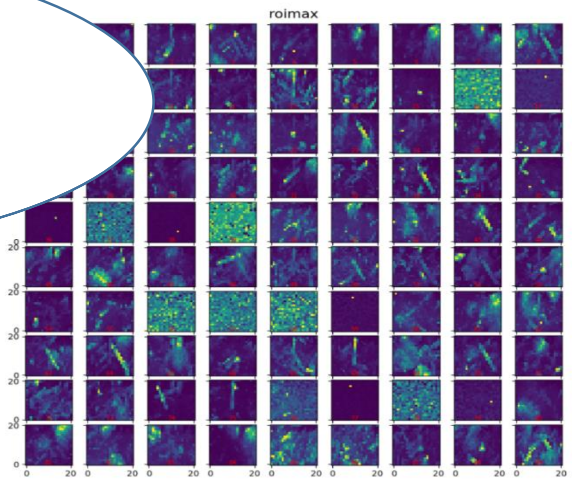
Low-carbon energy



UO_2

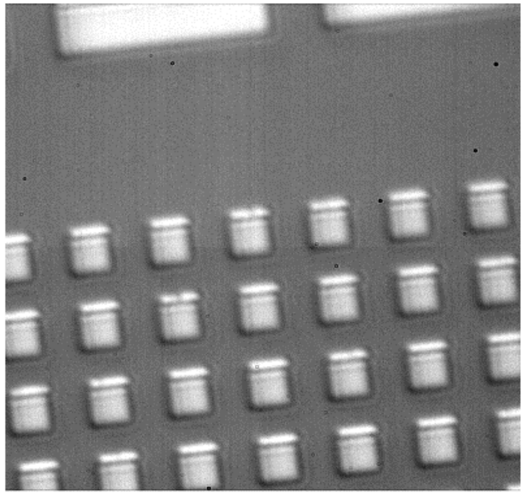


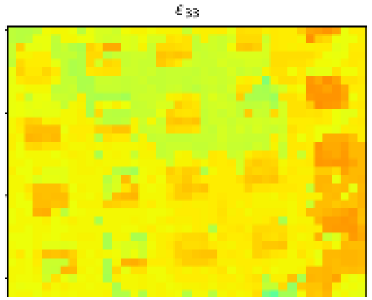
U-Zr-O (corium)

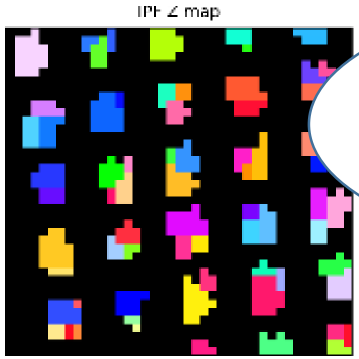


new battery materials

Applications

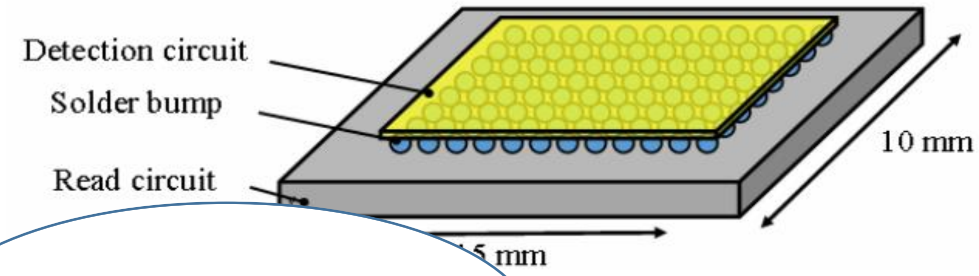






3D interconnects

circuits solder



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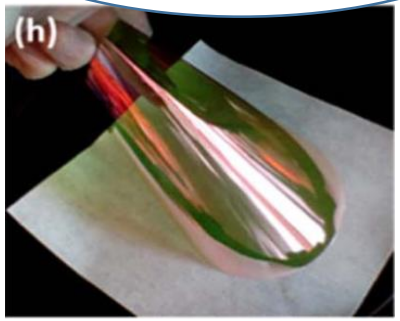
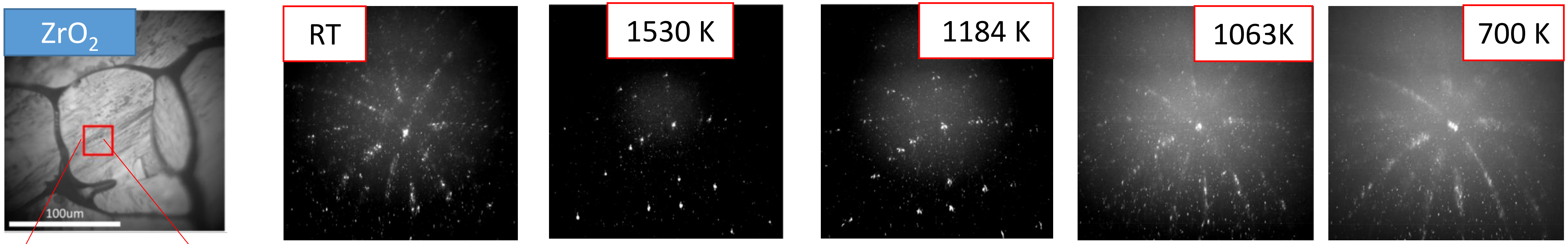
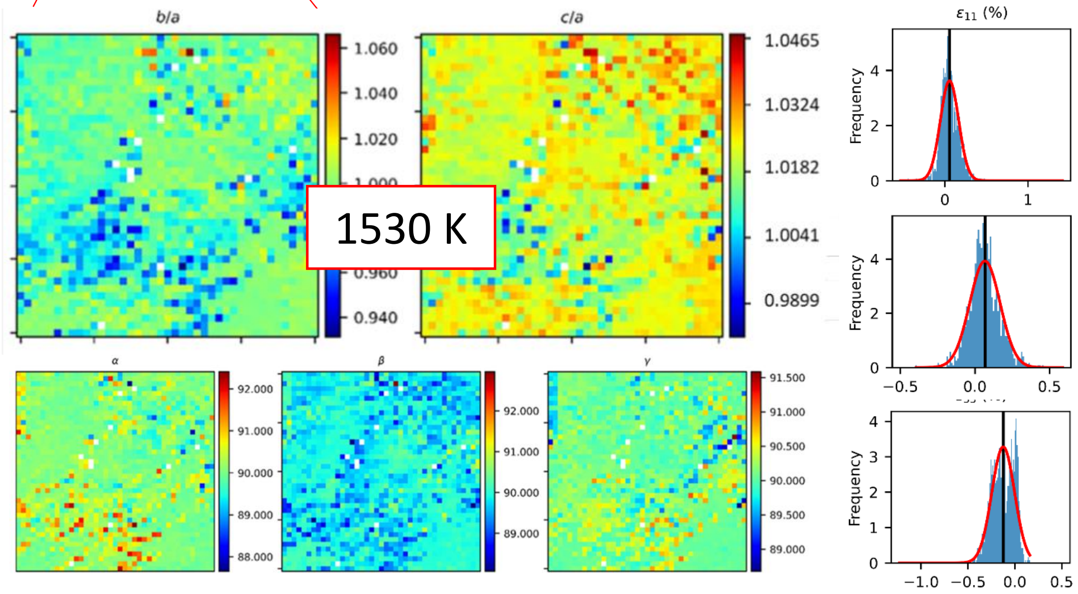
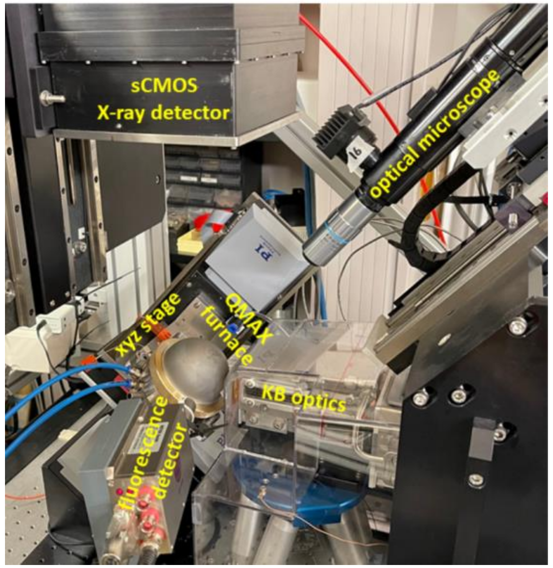
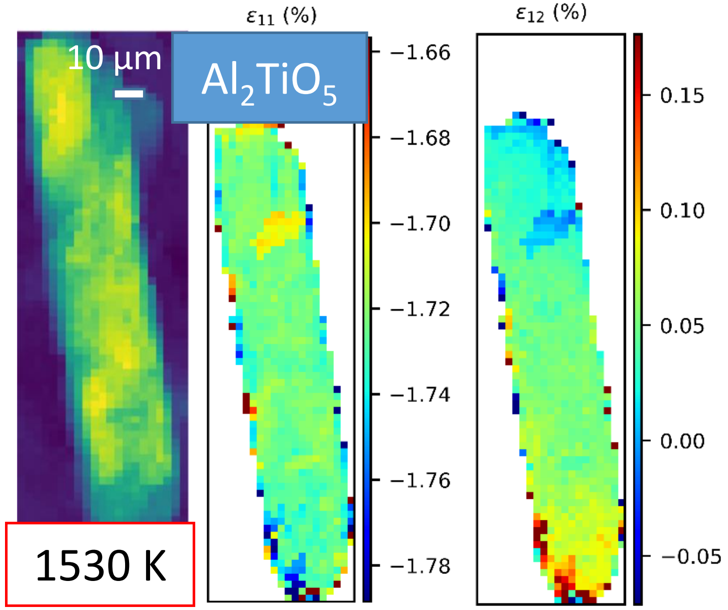


Flexible microelectronics



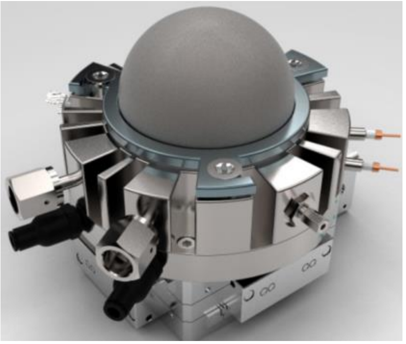






Collab. R. Guinebretière's group IRCER, Limoges, HotMIX project)

Ceramic paste/Pt heater resistor
50 K/min
PEEK, Be Dome
Air, O_2, vacuum



Furnace

R. Guinebretière et al, J. Appl. Cryst. (2022)

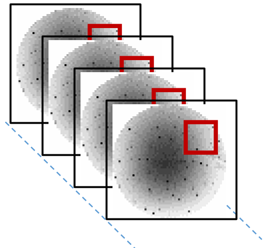
Experimental premiere >1500K , calibration without microscope

R.R.P. Purushottam Raj Purohit et al, submitted to J. Appl. Cryst.

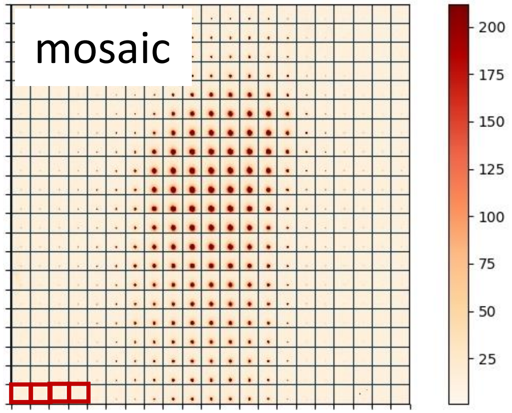
Strain distribution evolution with T

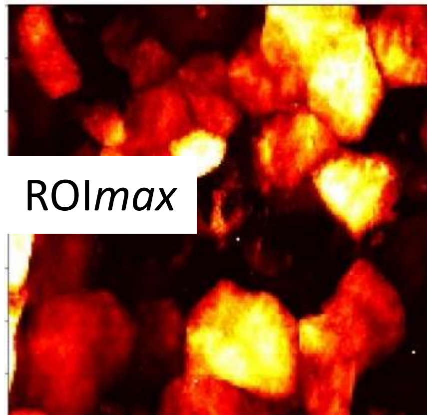
D. Fowan et al, in preparation

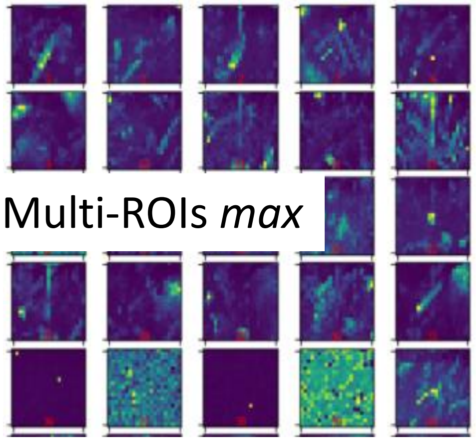
Microscopy Imaging without indexing



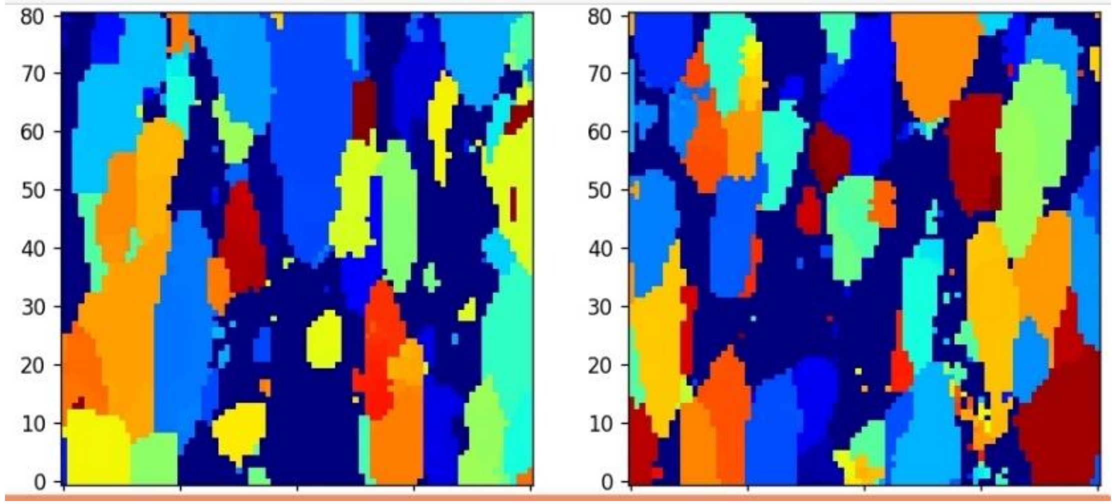
X ray detector: ROI counters
- *2D: Mosaic*
- *Scalar: max, mean, maxpos*





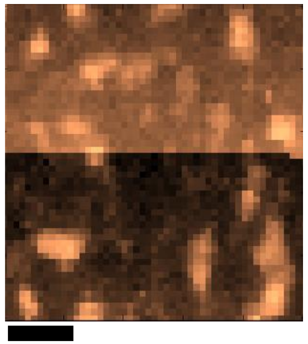


Peaks lists dataset
peak existence, correlation, factorization,…

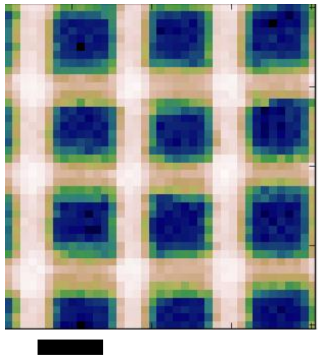


Multi-ROIs *maxpos*

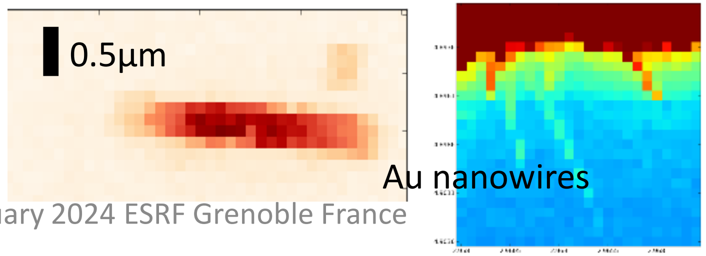
Fluorescence map



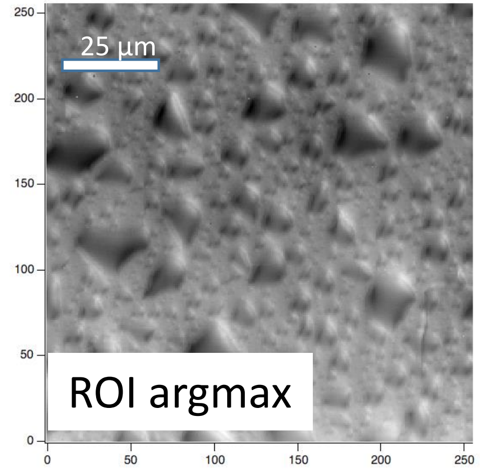
10µm NdFeB



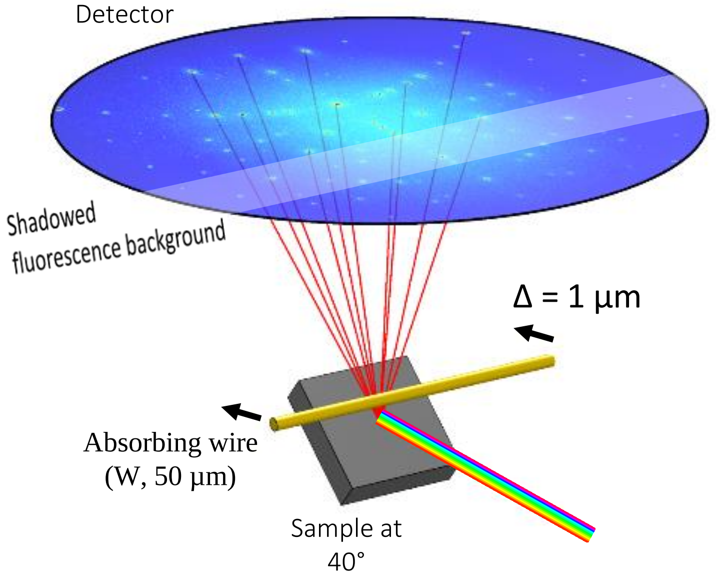
2µm Cu pads/Si

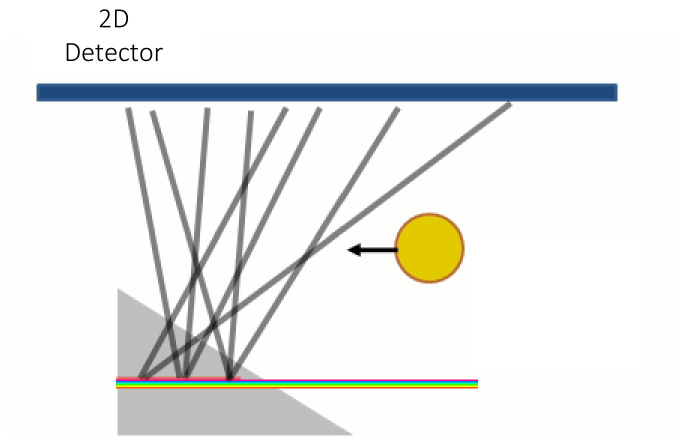


Au nanowires

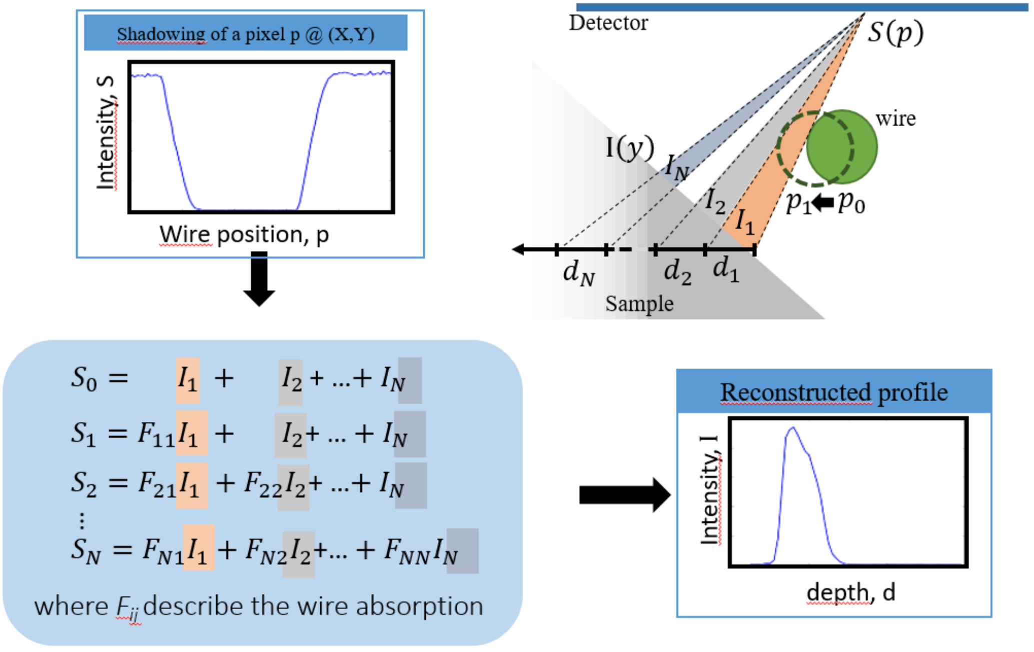


3D Laue Microscopy: principles





Improvements:
- Transmission function of wire
- 3-5 wires in parallel

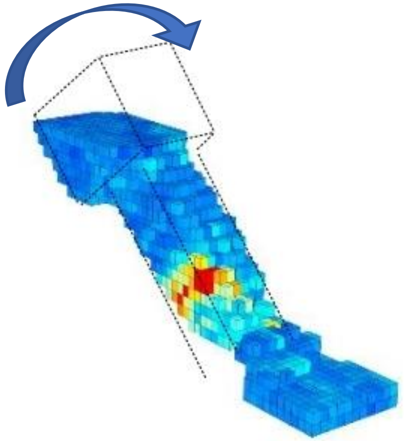


- ➢ Penetration depth from µm to mm
- ➢ DAXM
  "Differential Aperture X-ray Microscopy"
  (Larson et al. 2002)
- ➢ Depth resolution: simple triangulation of the scattered intensity

Collab. C. Kirchlechner's group (MPIE, KIT), Germany, XmicroFatigue project

3D-µLaue : data collection and analysis

In situ plasticity
fatigue mechanical test
Cu bicrystal

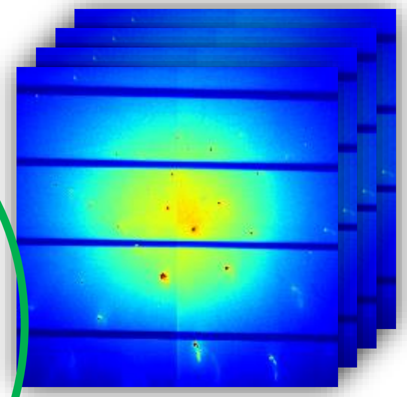


Damage and defects (dislocations)
Storage and interactions with surface and grain boundary

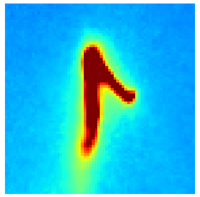
Local
Orientation
strain

$$\forall \text{ voxel}(x, y, z) \quad \begin{Bmatrix} p \\ \boldsymbol{U} \\ \boldsymbol{\varepsilon}^D \end{Bmatrix}$$

3D Scan (x, y, p)

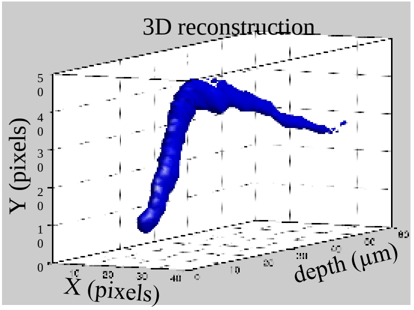


Segment



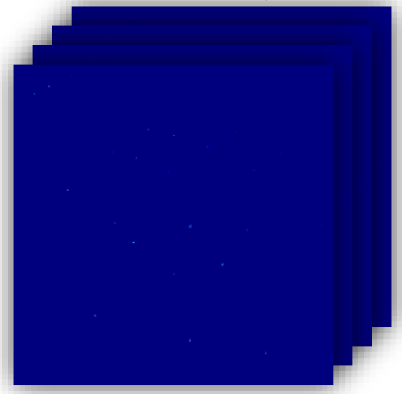
Reconstruct

✓ New software package included in LaueTools



Assemble

3D map (x, y, z)



Laue analysis

Local Laue pattern p
$\forall$ voxel(x, y, z)

Collab. C. Kirchlechner's group (MPIE, KIT), Germany, XmicroFatigue project

Energy measurements: full strain/stress tensors

Full strain

2 measurements

Deviatoric strain

$$\varepsilon^* = \varepsilon - \bar{\varepsilon} I_3$$

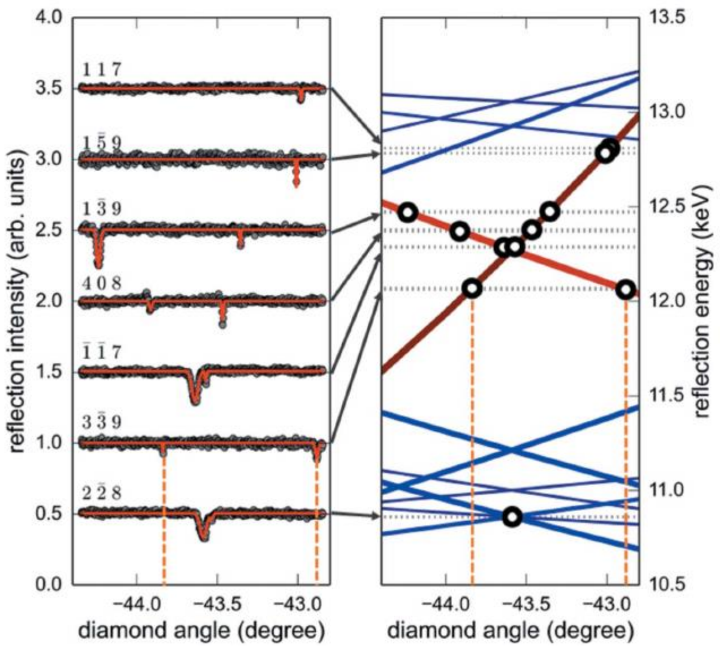
hydrostatic strain

hkl Laue spot energy (dspacing)

$$\epsilon_{hkl} = \bar{\epsilon} + (\epsilon^* \tilde{\mathbf{G}}).\tilde{\mathbf{G}}$$

Full stress

$\sigma = C\varepsilon$



Rotating diamond transmission filter

Energy resolution:

Intrinsic HR from crystal diffraction (monochromator, crystal analyser): some 10^{-4}

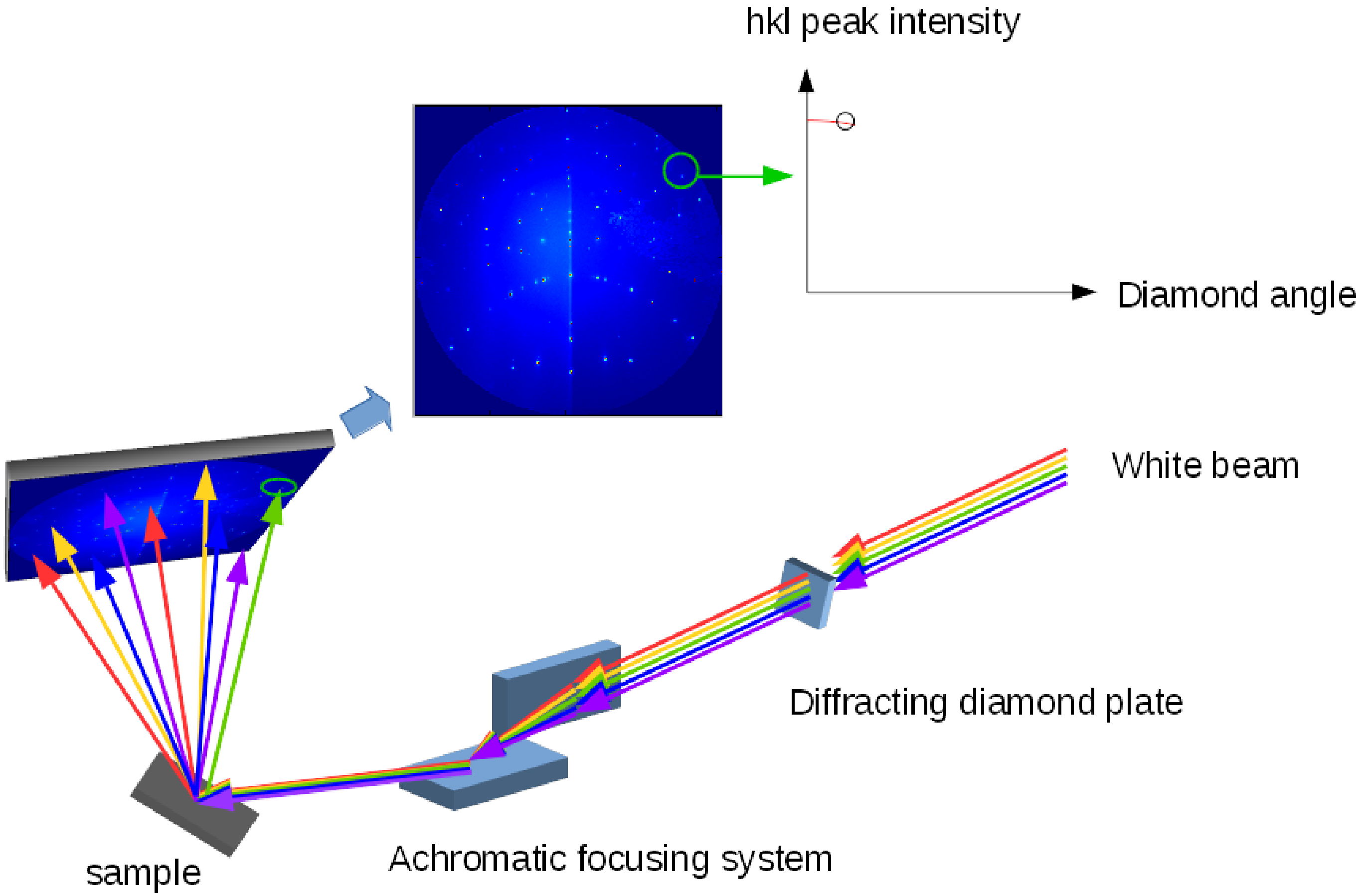
Much Less resolution for solid state detector (punctual SDD, 2D pnCCD): ~ 10^{-2} (broadening)

< 10-3 (average energy dsitribution)

HR from dev. Strain by assuming $\sigma_{zz} = 0$ (at surface). Suited for Thin film 2D map

µLaue Diffraction | energy fine measurements using rotating transmission diamond filter | Full Stress

"Inverse" monochromatic diffraction **The Rainbow filter technique**



Robach *et al.*, Acta Cryst. (2013). A**69**, 164-170

µLaue Diffraction | energy fine measurements using rotating transmission diamond filter | Full Stress

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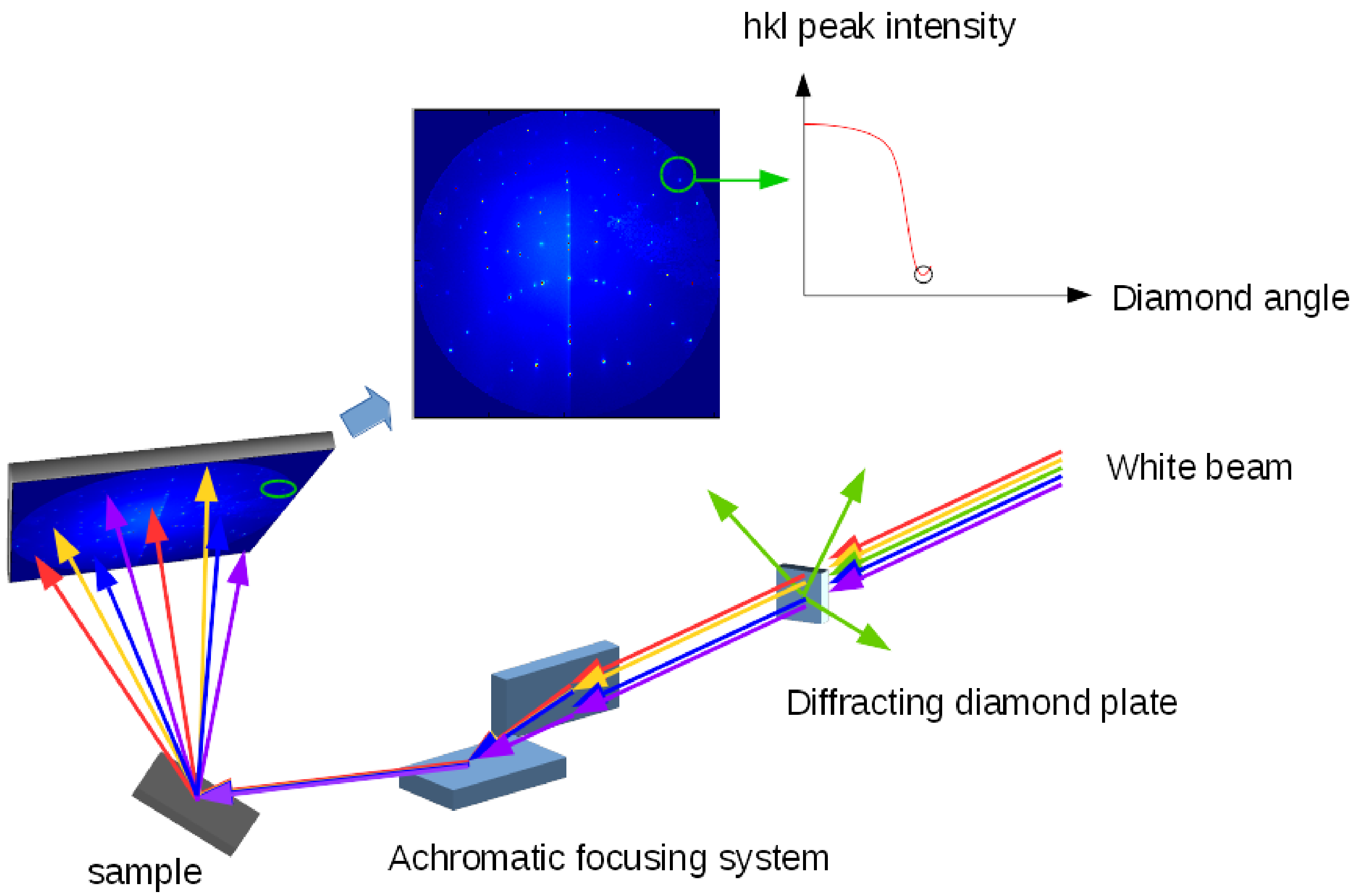

Robach *et al.*, Acta Cryst. (2013). A**69**, 164-170

µLaue Diffraction | energy fine measurements using rotating transmission diamond filter | Full Stress

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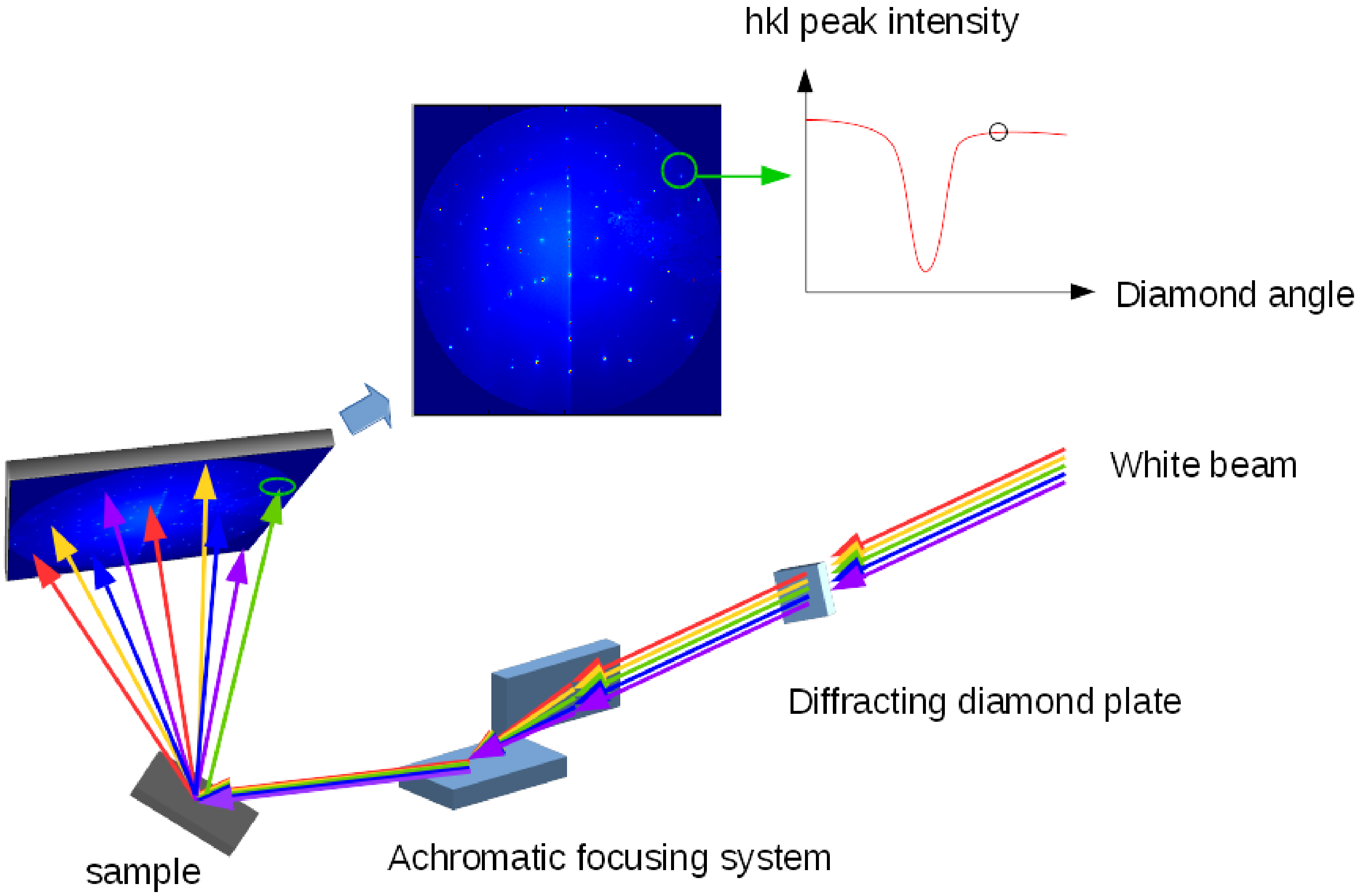

Robach *et al.*, Acta Cryst. (2013). A**69**, 164-170

Conclusions µLaue @ ESRF

Microstructure and Metrology characterisation tool

- ✓ **Open to broad scientific community**
- ✓ High angular resolution
- ✓ Few 100s nm spatial resolution
- ✓ *In situ* experiments (elasticity – plasticity), signal with large strain
- ✓ Addons mapping measurements: 3D, full strain/stress, element concentration

Next proposals call: March42023
Contact | infos | free tests
micha@esrf.fr

Current & future challenges

- ✓ LaueMAX Upgraded instrument (flux X 10, beamsize 150 nm) coming soon Spring 2024
- ✓ use of AI assistance (DIADEM project): - data collection (detect and select high quality data)
  - data analysis (peaks overlap, peaks subgrains splitting)

Crowded Laue patterns
(multiple grains, twins)
Peak Shape analysis
(Defects)
Infer topography
(depth, GB, 3D shape)